

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In Re Application of)	For: LOW PRESSURE REVERSE
)	OSMOSIS AND NANOFILTRATION
JOHN EDWARD TOMASCHKE)	MEMBRANES AND METHOD FOR
)	THE PRODUCTION THEREOF
Serial No.: 09/724,883)	
)	
Filing Date: November 28, 2000)	Group Art Unit: 1723

**SUBSTITUTE BRIEF ON APPEAL UNDER 37 C.F.R. §§ 41.31 and 41.37;
REFERRAL TO CANCELLATION OF CLAIMS UNDER 37 C.F.R. § 41.33**

Mail Stop Appeal Brief - Patents
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Attention: Krishnan S. Menon
Examiner

Dear Sir:

This substitute Appeal Brief is filed under the provisions of 35 U.S.C. § 134 and 37 C.F.R. Part 41, specifically §§ 41.31 and 41.37, and in response to the Office Action of March 29, 2005. A Notice of Appeal was filed on in this case on January 24, 2005. Enclosed herewith are a) an Amendment to cancel certain of the claims previously noticed as being appealed and b) a marked-up copy of the original Appeal Brief to indicate the changes made thereto in response to the Office Action.

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on April 27, 2005.

Linda Farrell

(Name)

(Signature)

April 27, 2005

(Date of Signature)

I. Real Party in Interest

The real party in interest on this appeal is Hydraunatics, Inc., a corporation and the assignee of Appellant Tomaschke.

II. Related Appeals and Interferences

There are no related appeals or interferences.

III. Status of Claims¹ and Identification of Claims on Appeal

Appellant's attorney has noticed that prior amendments have resulted in independent Claims 15 and 29 inadvertently being made identical. As the sets of dependent claims following each independent claim are parallel, that has resulted in the claims on appeal being two duplicate sets.

Appellant is therefore submitting herewith an Amendment to cancel Claims 29-39 inclusive as duplicative. While Appellant agrees with the Examiner's statement in the Office Action of March 29, 2005, that amendment is not a matter or right, Appellant notes that appellate review of two duplicate sets of claims is counterproductive and will therefore presume -- hopefully correctly -- that the amendment will be approved. Therefore Appellant will proceed in this brief with arguments directed to appeal of Claims 15-21, 23, 25 and 27-28, the remaining claims not previously cancelled or withdrawn, and will not expressly address Claims 29-39 other than as required by the Patent Office Rules.

Should Appellant have made such presumption incorrectly and the accompanying Amendment is not accepted or entered, then Appellant respectfully requests that the Board of Appeals consider that all arguments made herein in support of patentability of Claims 15-21, 23, 25 and 27-28 and reversal of the rejections thereof

¹ The original claims were numbered 1-13 and 15-27. The number "14" in the original claim sequence was inadvertently omitted. In the first Office Action the Examiner renumbered Claims 15-27 as Claims 14-26, and then issued a restriction requirement in which renumbered Claims 1-14 were defined as Group I and Claims 15-26 were defined as Group II. The claims of Group II (Claims 15-26) were elected and remained in the present application while Claims 1-14 were cancelled as being directed to a non-elected invention. The status of Claims 13 and 14 as product-by-process claims was considered and the Examiner ruled that those claims properly belonged to Group I.

are also fully and equally applicable with respect to Claims 29-39 and reversal of the rejections thereof, and render a decision on appeal as to all Claims 15-21, 23, 25 and 27-39 accordingly.

Appellant's attorney apologizes for any inconvenience that this may have caused the Examiner or the Board of Appeals.

a. Identification of Claims on Appeal:

The claims on appeal are Claims 15-21, 23, 25 and 27-39, inclusive, prior to entry of the accompanying Amendment and Claims 15-21, 23, 25 and 27-28, if said Amendment is entered.

b. Status of All Claims in the Proceeding:

Prior to Amendment:

Claims 1-14:	Cancelled
Claims 15-21:	ON APPEAL; rejected
Claim 22:	Cancelled)
Claim 23:	ON APPEAL; rejected
Claim 24:	Cancelled
Claim 25:	ON APPEAL; rejected
Claim 26:	Cancelled
Claims 27-39:	ON APPEAL; rejected

Following entry of Amendment:

Claims 1-14:	Cancelled
Claims 15-21:	ON APPEAL; rejected
Claim 22:	Cancelled)
Claim 23:	ON APPEAL; rejected
Claim 24:	Cancelled
Claim 25:	ON APPEAL; rejected
Claim 26:	Cancelled
Claims 27-28:	ON APPEAL; rejected
Claims 29-39:	Cancelled

IV. Status of Amendments

Other than the amendment made herewith under 37 C.F.R. § 41.33 to cancel Claims 29-39, there are no amendments which have been submitted since the final rejection and/or have not been entered.

V. Summary of Claimed Subject Matter

The subject matter of the invention is a composite membrane commonly referred to as a reverse osmosis ("RO") or nanofiltration ("NF") membrane for water purification. More particularly, the composite membrane is defined in Claim 15 as:

A composite membrane (§ 0007, pg. 3) useful for reverse osmosis or nanofiltration (§ 0003, pg. 1) comprising:

a supportive porous under-structure (§ 0007, pg. 3); and
adhered thereto, a top layer consisting of a crosslinked polyamide thin film (§ 0007, pg. 3; § 0009, pg. 3) which has been contacted with a solution of a C_1 - C_6 alkyl, alkenyl, haloalkyl, haloalkenyl, or hydroxy sulfonic or disulfonic acid compound (§ 0010, pg. 3; § 0011, pg. 4; original Claim 23),

whereby the resulting membrane has a water flux of at least about 15 gallons per square foot per day ("gfd") and a rejection of at least 20 percent when tested with 0.05% aqueous sodium chloride at 75 psi and 25°C (§ 0003, pg. 2).

The most important sulfonic acid compounds from a commercial standpoint are methanesulfonic acid ("MSA"; § 0011, pg. 4; § 0017, pg. 6; § 0022, pg. 7; original Claim 23) and trifluoromethanesulfonic acid ("TFMSA"; § 0024, pg. 8; original Claim 23). Also of important is ethanesulfonic acid ("ESA"; amended Claim 27), the analog of MSA.

The critical element in this invention which produces a superior membrane is that the top layer of the membrane has been *contacted with the low molecular weight (C_1 - C_6) sulfonic or disulfonic acid* (§ 0003, pp 1-2). A claimed membrane so treated requires *no other treatment* involving rejection enhancing agents to obtain the superior rejection and flux properties defined in the claims (§ 0003, pg. 2).

As will be shown below, and as has been fully established in the record of the application by comparative and conclusive data, no other single treatment method can achieve this result. While the mechanism is not known with certainty, it is believed that these the application of these sulfonic acids alters the physical characteristics of the

polyamide top layer of the membrane by swelling the crosslinked polyamide and solvating the uncrosslinked polyamide structures, so that subsequent rinsing removes the solvated fragments from the crosslinked film matrix leaving being a more permeable polymeric skin layer with the enhanced water permeability and salt rejection properties (¶ 0010, pp. 3-4).

The actual treatment procedure is described in ¶ 0015 (pg. 5) of the Specification, and involves application of about 1-15 gms of acid per m² of membrane surface, followed by drying (optionally with mild heating). Duration and temperature of acid application and drying will depend on the specific sulfonic acid being used and the desired degree of permeability to be obtained.²

VI. Grounds of Rejection to be Reviewed on Appeal

The Final Rejection in the Office Action of August 11, 2004 (hereinafter referred to as "FR") states two grounds of rejection, both under 35 U.S.C. § 103(a). Both are on appeal and they are related.

- A. Claims 15-20 and 25 have been rejected under § 103(a) as obvious over:
 - a) Chau et al., U.S. Patent No. 4,983,291, in view of
 - b) 1) "applicant's [Appellant's] own admission by 131³ affidavit" and
 - 2) Kiefer et al., "OPTIMIZING THE PERFORMANCE OF LOW FOULING MEMBRANES FOR THE WORLD'S LARGEST NANOFILTRATION PLANT," a technical paper presented at the American Water Works Association meeting in Boca Raton, Florida, held March 2-5, 2003.

(FR, ¶ 1, pp. 2-3)

B. Claims 21, 23 and 27 have also been rejected under 35 U.S.C. § 103(a), based on Chau et al. in view of "applicant's admission" and Keifer et al, and *further in view of* Koo et al., U.S. Patent No. 6,063,278 . (FR, § 2, pp.3-4)

² As noted above, original Claims 1-14 related to the method of manufacture of the claimed membrane were subject to restriction and cancelled as non-elected, so the method of manufacture is described here only for information and is not subject to the present appeal.

³ The Examiner in his final rejection refers to Appellant's so-called "admission" as being made in a "131 Affidavit." In fact Appellant has not filed any Affidavit under 37 C.F.R. ' 1.131. Rather Appellant's evidence statements to rebut prior art have been in the nature of affidavits filed under 37 C.F.R. ' 1.132. Appellant herein assumes that the Examiner's reference to a "131 Affidavit" is inadvertent, and the arguments herein are based on that assumption.

Claim 28, which is limited to the specific use of TFMSA as the sulfonic acid, has been objected to but is deemed allowable if rewritten in independent form including all limitations of the preceding claims. (FR, "Allowable Subject Matter", pp. 4-5)

For both rejections, it is the Examiner's contention that Chau et al. discloses an equivalent membrane which has been contacted with a "sulfonic acid compound" and which has equivalent properties of salt rejection and flux (FR, ¶ 1, pg. 2). The Examiner contends that Chau et al discloses "sulfonic acid, toluene sulfonic acid, amine salt of sulfonic acid" as contacting agents (FR, ¶ 1, pg. 2). He acknowledges that Chau et al. does not teach whether other sulfonic acids would have the same effect, but he contends that "applicant's admission" and Keifer et al. provide such teaching (FR, ¶ 1, pg. 3).

For the second rejection, he contends that Koo et al. discloses MSA, ESA and benzene sulfonic acid for making RO membranes and that it would be obvious to one skilled in the art to use Koo et al's teaching in Chau et al's system (FR, ¶ 2, pg. 4).

The Examiner's reason for deeming two claims deemed allowable is that in his view TFMSA is not taught by either Chau et al. or Koo et al. and that Appellant's data show superior results for TFMSA that is not shown for other sulfonic acids (FR, pg. 5).

VII. Arguments

Appellant submits that there are several reasons why the Examiner's rejections are not well founded and why Appellant's invention would not be obvious to one skilled in the art under § 103(a).

1. From a chemistry standpoint, all of the following are applicable to some degree:

A. Chau et al. and Koo et al. do not teach membranes treated by sulfonic acids as claimed by Appellant;

B. The prior art disclosures of use of sulfonic acids by Chau et al. and Koo et al. in membrane production involve uses of sulfonic acids other than treating for property enhancement or the sulfonic compounds disclosed for treatment are not sulfonic acids or the equivalent thereof.

C. Neither Chau et al. nor Koo et al. teaches or suggests any enhancement of membrane properties of the type present in Appellant's membranes.

2. From a procedural and legal standpoint,

A. Appellant's statements submitted in the present application do not contain any adverse "admission" of facts as contended by the Examiner; rather Appellant's statements all support a clear distinction between his invention and those of Chau et al. and Koo et al.

B. The Keifer et al. article was not published or presented until 2003 and therefore also is not prior art under § 103(a) against Appellant's application, which was filed in 2000.

Therefore, as Appellant will establish herein:

1. Both of the Examiner's combination rejections rely upon a) the Keifer et al. reference, which was not available prior to Appellant's filing date, and b) an alleged "admission" which does not exist, such that neither is properly part of the final rejection. Since the Examiner has not made any other rejection, he has acknowledged that Appellant's claims cannot be rejected without reliance on those two components. Therefore the two remaining components of the combinations (Chau et al. in the first rejection and Chau et al. plus Koo et al. in the second rejection) are by the Examiner's own action acknowledged to be insufficient to reject Appellant's invention under the requirements of § 103(a); and

2. Since the Examiner's combination of the two referenced patents (Chau et al. and Koo et al.) does not teach Appellant's claimed membranes or their chemistry or refute Appellant's data showing the clear superiority of the claimed membranes in a manner which would be considered by one skilled in the art to be relevant, the rejections also do not meet the requirements of § 103(a).

a. Critical cited references are not prior art:

There is no question but that patent or publication references which are cited to support a rejection under §§102 or 103(a) must have been publicly available prior to the filing date of an Appellant's application; see *In re Koller et al.*, 613 F.2d 819, 204 U.S.P.Q. 702 (C.C.P.A.: 1980)⁴.

⁴ The only exceptions are where a patent reference meets the requirements of 35 U.S.C. § 102(e), which applies to Koo et al., or where a post-filing reference is cited to show pre-filing facts, which is not

The present application was filed in November 2000. In his first rejection, two of the three references which the Examiner includes in his combination of alleged "prior art" -- Appellant's so-called "admission" and the Keifer et al. paper -- did not exist until at least 2003⁵. They therefore cannot be incorporated into a combination rejection of Appellant's claims.

Similarly, In his second rejection, those two references make up two of the four references of the cited combination, notwithstanding that here they cannot be incorporated into a combination rejection of Appellant's claims.

Thus, neither of the Examiner's grounds of rejection is proper under § 103(a), since both of the combinations of "prior art" relied upon include documents or alleged statements which did not exist prior to Appellant's filing date. *Therefore on this ground alone both of the Examiner's rejections must be reversed.*

It must be accepted that if the Examiner had believed that a rejection was possible without reliance on the two post-2000 references, he would have made such a rejection. He has not done so. It is significant that these rejections came only after Appellant had submitted extensive comparative data refuting the Examiner's prior rejections and clearly establishing the major differences between the chemistry and properties of Appellant's membranes and method as compared to those of the previously cited Chau et al and Koo et al. The fact that the Examiner then found himself forced to incorporate the two post-2000 references into his rejections in reply to those data necessarily means that he considers those two post-2000 references to be critical to his rejection of all of Appellant's claims. Thus it is not now possible for either of his rejections to be upheld, since the presence of those post-2000 references expressly violates the requirements of § 103(a).

Removal of those two references leaves only Chau et al. as the basis for the Examiner's first rejection, and the Examiner makes no contention that Chau et al. is in

applicable in the present case.

⁵ Applicant has actually submitted two 132 Affidavits, one in 2003 and the other in 2004 (and an Informal Statement in the nature of a 132 Affidavit in 2004); see Part IX: Evidence Appendix below. The Examiner has not indicated specifically which document he is referring to. However, that is of no consequence in this case, since all have post-dated the filing of this application in 2000 and therefore none can be prior art.

any manner sufficient in and of itself. Indeed, it will be noted that the Examiner in the subject final rejection dropped prior rejections based on other references and thus acknowledged that only with the inclusion of the "admission" and Keifer et al. could basis for a rejection be found. Thus the rejection of Claims 15-20 and 25 over Chau et al. *in view of "Appellant's admission" and Keifer et al.* therefore must necessarily fall without the two secondary references.

The second rejection fares no better. With the "admission" and Keifer et al. gone, supplementation of Chau et al. with Koo et al. does not overcome the fatal deficiencies of the isolated Chau et al. reference. The Examiner's acknowledgement that Chau et al. alone is insufficient, coupled with his citation of Koo et al. as being of only limited relevance, makes it clear that the rejections of Claims 21, 23 and 27 under § 103(a) also cannot stand in the absence of the two post-2000 references.

Reversal of both of the Examiner's grounds of rejection is therefore respectfully requested.

b. The Chau et al. and Koo et al. disclosures do not make Appellant's invention obvious under § 103(a)

In addition to the lack of a supportable combination of references as discussed above, it is clear that the disclosures of Chau et al. and Koo et al. do not describe membrane technology which would be considered by one skilled in the art to in any way teach or suggest Appellant's claimed membranes within the requirements of § 103(a).

Considering first Chau, Chau is a broad but quite unspecific disclosure of acid treating of membranes. Chau has a long list encompassing numerous classes of acids and amines, including classes as large as the "inorganic acids" (col. 3, lines 45-48 and 62-65; col. 6, lines 60-65). Of these "sulfonic acids" is only one of the many classes. Only two specific sulfonic acids are identified (p-toluene sulfonic acid and m-toluene sulfonic acid; col. 7, lines 9-10)⁶, and neither of these is a C₁-C₆ sulfonic acid. Further, it is clear that the sulfonic acid class is not favored by Chau. Only a single sulfonic acid

⁶ The reference in that paragraph on line 8 to "sulfonic acid" is of no significance, since there is no recognized compound named "sulfonic acid." See, e.g., Grant, HACHK'S CHEMICAL DICTIONARY (4th ed.), p. 646 (McGraw-Hill Co., 1969 and Lewis, HAWLEY'S CONDENSED CHEMICAL DICTIONARY (13th ed.), p. 1060 (John Wiley & Sons, 1997). A rejection of a chemical invention cannot be based on a mention of a non-existent material.

(p-toluene sulfonic acid: "PTSA") appears in any of the Examples (Example 5, Table 4) and its properties are decidedly inferior to many other compounds tested, most notably the citric acids of Example 3, Table 2. It is clear, therefore, that no specific sulfonic acid meeting Appellant's claims is disclosed in the Chau patent.

It is well settled that a generic disclosure of a chemical class is not necessarily a disclosure of the individual members of the class. The disclosure must be assessed to determine whether one skilled in the art would be taught the particulars of the claimed invention, involving only a portion of that class, from the generic disclosure. In the present case, the teaching of Chau is away from such understanding by the person skilled in the art. Not only are the sulfonic acids as a class taught as inferior to other acids by Chau and the exemplification by Chau of individual sulfonic acids do not include any examples within the scope of Appellant's claims, but in addition the only example of the sulfonic acid tested by Chau (PTSA) is shown to have properties outside the required properties of sulfonic acids for Appellant's invention. Specifically, in Chau's Table 4, PTSA is shown to have a flux some 10% less than the minimum required in Appellant's claims. This is evidenced in the current record by the calculations presented to the Examiner in Appendix B of the Amendment submitted on May 7, 2004. Further, both p- and m-toluene sulfonic acid are solids at the contacting temperatures of Appellant's invention and therefore are inoperative for Appellant's claimed invention, since they cannot effectively contact Appellant's membranes to impart the claimed membrane properties.

Appellant has presented data in the Specification and in responses filed January 14, 2003 and May 7, 2004, which clearly establish the critical limitation that the C₁-C₆ sulfonic acids are those operable in the claimed invention. Nothing in the Chau reference discloses any such criticality or even mentioned this subclass of sulfonic acids. Appellant has found nothing -- nor has the Examiner indicated anything -- in the Chau reference which would suggest to one skilled in the art that one must have these particular sulfonic acids to obtain the superior properties of Appellant's membranes.

The Examiner acknowledges that Chau alone has insufficient disclosure to reject Claims 15-20 and 25 under § 103(a), since he has found it necessary to add the so-called "admission" and the Kiefer paper to obtain even minimal support for his rejection.

Since as noted above the "admission" and the Kiefer paper are not prior art. Therefore, in considering the rejection of those claims, only the Chau reference alone can be used to support the rejection, since the Examiner has not identified any other prior art document to supplement Chau. Chau, however, teaches nothing about Appellant's invention as claimed, and what Chau does teach about sulfonic acids at all is quite limited, negative and directed entirely away from Appellant's invention.

Therefore, since Chau cannot by itself support the rejection of Claims 15-20 and 25 under § 103(a) that rejection must be reversed, and such **reversal** is respectfully requested.

The Examiner has supplemented the rejection under § 103(a) of Claims 21, 23 and 27 over Chau with Koo as a tertiary reference, using the "admission" and Keifer as secondary references. Since the latter two are not prior art, the rejection must be considered to be one of "Chau in view of Koo." This rejection, also, cannot stand.

The Examiner's acknowledgement that Chau and Koo in combination are not sufficient to support the rejection has been discussed above, in connection with the impropriety of the "admission" and Keifer paper as prior art.

The rejection is also without merit even if one considers *arguendo* that the combination of Chau and Koo might stand even without the secondary references. The Examiner contends that these taken together teach Appellant's invention. Chau, however, is fatally deficient in not teaching anything about sulfonic acids as claimed in Appellant's claims, as discussed above. Koo not only does not overcome that deficiency, but in fact Koo does not even teach about sulfonic acids for treating membranes at all. Therefore not only does Koo not supplement Chau, in fact teaches even further away from Appellant's invention.

Koo does disclose sulfonic acids, including two of the C₁-C₆ sulfonic acids, methanesulfonic acid and ethanesulfonic acid (col. 6, lines 2-3), as well as many other "strong" acids such as nitric and hydrochloric acids. However, in Koo's process, none of the acids, including the sulfonic acids, are used for the production or treatment of membranes. Rather Koo uses amines or amine salts (col. 2, line 65-col. 3, line 5). The salts are the reaction products of strong acids and amines (col. 3, lines 6-7). Clearly a disclosure of production of a membrane from an amine or an amine salt does not teach

one skilled in the art anything about treatment of an membrane with a specific subgroup of sulfonic acids to enhance the membrane properties.

Since Koo does include low molecular weight sulfonic acids as among his "strong acids," Koo does disclose that those sulfonic acids can be reacted with amines to produce some of his amine salts. However, those skilled in the art would be aware from basic chemistry that a salt made from an acid is an entirely different compound with entirely different properties. The person skilled in the art would also be aware that such a salt reacts with other materials in ways that cannot be reliably predicted from the knowledge of the acid from which the salt was formed. Thus Koo's teaching of using amine salts made from sulfonic acids in the production of membranes can teach nothing to the person skilled in the art about how membranes as claimed in Appellant's claims would react to contact with the sulfonic acids. Indeed the teaching of Koo is further deficient in this area, since according to his process by the time the membrane is formed all acids are gone from his system, having been fully reacted into amine salts. Thus while sulfonic acids may be present at the beginning of Koo's membrane production method, they are quickly all reacted into the amine salts and do not further participate in any aspect of Koo's invention.

This disclosure makes Chau and Koo mutually incompatible and mutually contradictory. Chau seeks to treat membranes at the end of production with acids, including some high molecular weight sulfonic acids. Koo seeks to destroy all acids at the beginning of his process to produce amine salts. No one skilled in the art would consider combination of two disclosures, one of which teaches to destroy the very compounds that the other process requires for its operation.

Appellant respectfully directs the attention of the Board to his Amendment filed on June 18, 2003, and particularly to Appellant's Declaration appended thereto. In that Declaration Appellant presents direct comparisons of his system to those of Chau and Koo. Those data clearly establish that Appellant's invention is substantially superior to either of Chau or Koo. It will be evident that since the combination of Chau and Koo proposed by the Examiner represents the forced overlay of two mutually incompatible systems, one of which essentially acting to destroy the acids that the other requires, the data also establish that Appellant's invention must be even further differentiated from

the Examiner's Chau/Koo combination.

Since Chau and Koo A) would be recognized by those skilled in the art as not being able to be combined as the Examiner proposes, B) even if so combined would be recognized by those skilled in the art as inoperable, and C) individually or in combination seen by those skilled in the art to result in a system far inferior to Appellant's claimed invention, it follows that the rejection of Claims 21, 23 and 27 under § 103(a) is not supported by Chau in view of Koo and therefore the rejection must fall. **Reversal** of the Examiner's rejection of those claims is therefore respectfully requested.

Since the so-called "admission" and the Keifer paper are not prior art, it is believed by Appellant that they do not require analysis in this brief. Therefore Appellant will here note only the following as to those items:

1. The Examiner contends that Appellant "admits" that an membrane denoted the "Koch" membrane in the Keifer paper is equivalent to Chau's membrane. Appellant presented evidence about the Chau reference by name in the June 18, 2003 response, and showed why his invention was far superior. Why the Examiner now thinks that merely mentioning the same membrane by another name is some sort of "admission" is not understood by Appellant, since the more recent submission by Appellant of the Keifer paper is merely a continuation of the arguments Appellant has been making in this case all along. Clearly neither the mention of the Koch membrane nor Appellant's data on the Chau membrane can in any way help the Examiner to overcome the fatal deficiencies in the Chau reference.
2. The Examiner also contends that Appellant "inadvertently" "admitted" that Chau's membranes' properties are superior. Appellant has countered that by providing calculations in Appendix B of the May 7, 2004 response to support his determination that the Chau membrane is inferior in its properties, contrary to the Examiner's contentions. The Examiner has not rebutted those calculations nor provided calculations of his own, such as by an Examiner's Affidavit under 37 C.F.R. § 1.104(d)(c), to support his contention.
3. In any event, Appellant notes that the Examiner's reliance on Appellant's Affidavit or on the Keifer mention of a Koch membrane is of no consequence, since the Affidavit was submitted during prosecution of this application and thus was not even in existence

prior to the filing date of the application, and the Keifer paper was not published until 2002, so by definition neither one can be relied upon or cited as "prior art".

Oral Hearing:

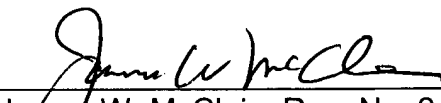
An oral hearing is not being requested.

Conclusion And Request

In view of the facts and arguments set forth above, Appellant submits that it has been shown conclusively that the Examiner's rejections are supported neither by fact nor by law, and that the rejections therefore are without merit and cannot stand. Appellant therefore respectfully requests that this Honorable Board of Appeal **REVERSE** the Examiner's ruling in total, rule that the rejections do not meet the requirements of 35 U.S.C. § 103(a) as to any of the rejected claims, and remand the application to the Examiner for allowance of all Claims 15-21, 23, 25 and 27-28, all claims currently in the application and on appeal herein.

Date Filed: March 8, 2005
Date Substituted: April 27, 2005

Respectfully submitted,


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Attorney's Docket No.: BHYNA-1028433

VIII: Appendix A: Claims on Appeal:

(Prior to entry of concurrently submitted Amendment; effect of such entry will be to reduce claims on appeal to Claims 15-21, 23, 25, and 27-28 inclusive)

15. A composite membrane useful for reverse osmosis or nanofiltration comprising:
a supportive porous under-structure; and
a top layer consisting of a crosslinked polyamide thin film which is adhered to the upper surface of the porous support structure, said top layer having been contacted with a solution of a C₁-C₆ alkyl, alkenyl, haloalkyl, haloalkenyl, or hydroxy sulfonic or disulfonic acid compound,

whereby said membrane has a water flux of at least about 15 gfd and a rejection of at least 20 percent when tested on a 0.05 percent aqueous sodium chloride at 75 psi and 25°C.

16. A composite membrane as in Claim 15 wherein said crosslinked polyamide comprises the reaction product of an aromatic diamine or triamine and an aromatic triacyl halide.

17. A composite membrane as in Claim 16 wherein said crosslinked polyamide comprises the reaction product of an aromatic diamine or triamine, an aromatic triacyl halide and an aromatic diacyl halide.

18. A composite membrane as in Claim 15 wherein the porous support is a polyarylethersulfone.

19. A composite membrane as in Claim 15 wherein said first membrane comprises a thin film, flat sheet, hollow fiber or tubular membrane.
20. A composite membrane as in Claim 15 wherein the membrane is a component of a spiral-wound membrane filter or a plate and frame filter.
21. A composite membrane as in Claim 15 wherein said organic sulfonic or disulfonic acid compound comprises a sulfoacetic, sulfosuccinic, methanesulfonic, ethanedisulfonic, or hydroxybutane sulfonic compound or mixtures thereof.
23. A composite membrane as in Claim 15 wherein said organic sulfonic acid compound comprises methanesulfonic acid, trifluoromethanesulfonic acid or a mixture thereof.
25. A composite membrane as in Claim 15 wherein said solution of an organic sulfonic acid compound comprises said organic sulfonic acid compound dispersed or dissolved in water, alcohol, glycol, alkoxy alcohol or a carboxylic acid or a mixture thereof.
27. A composite membrane as in Claim 15 wherein said organic sulfonic acid compound comprises ethanesulfonic acid.

28. A composite membrane as in Claim 15 wherein said organic sulfonic acid compound comprises trifluoromethanesulfonic acid.

29. A composite membrane useful for reverse osmosis or nanofiltration comprising:
a supportive porous under-structure; and
a top layer consisting of a crosslinked polyamide thin film which is adhered to the upper surface of the porous support structure, said top layer having been contacted with a solution of a C₁-C₆ alkyl, alkenyl, haloalkyl, haloalkenyl, or hydroxy sulfonic or disulfonic acid compound,

whereby said membrane has a water flux of at least about 15 gfd and a rejection of at least 20 percent when tested on a 0.05 percent aqueous sodium chloride at 75 psi and 25°C.

30. A composite membrane as in Claim 29 wherein said crosslinked polyamide comprises the reaction product of an aromatic diamine or triamine and an aromatic triacyl halide.

31. A composite membrane as in Claim 30 wherein said crosslinked polyamide comprises the reaction product of an aromatic diamine or triamine, an aromatic triacyl halide and an aromatic diacyl halide.

32. A composite membrane as in Claim 29 wherein the porous support is a polyarylethersulfone.

33. A composite membrane as in Claim 29 wherein said first membrane comprises a thin film, flat sheet, hollow fiber or tubular membrane.

34. A composite membrane as in Claim 29 wherein the membrane is a component of a spiral-wound membrane filter or a plate and frame filter.

35. A composite membrane as in Claim 29 wherein said organic sulfonic or disulfonic acid compound comprises a sulfoacetic, sulfosuccinic, methanesulfonic, ethanedisulfonic, or hydroxybutane sulfonic compound or mixtures thereof.

36. A composite membrane as in Claim 29 wherein said organic sulfonic acid compound comprises methanesulfonic acid, trifluoromethanesulfonic acid or a mixture thereof.

37. A composite membrane as in Claim 29 wherein said solution of an organic sulfonic acid compound comprises said organic sulfonic acid compound dispersed or dissolved in water, alcohol, glycol, alkoxy alcohol or a carboxylic acid or a mixture thereof.

38. A composite membrane as in Claim 29 wherein said organic sulfonic acid compound comprises ethanesulfonic acid.

39. A composite membrane as in Claim 29 wherein said organic sulfonic acid compound comprises trifluoromethanesulfonic acid.

IX: Appendix B: Evidence:

Attached hereto are copies of the following evidence documents which have been entered into the file of this application during the prosecution thereof by Appellant or the Examiner and which are mentioned in Appellant's arguments above:

A. Evidence submitted by Appellant:

Exhibit 1: Kiefer et al., "OPTIMIZING THE PERFORMANCE OF LOW FOULING MEMBRANES FOR THE WORLD'S LARGEST NANOFILTRATION PLANT," a technical paper presented at the American Water Works Association meeting in Boca Raton, Florida, held March 2-5, 2003; entered into record by Appellant as Appendix D to Appellant's Amendment filed May 7, 2004.

Exhibit 2: Declaration of John. Tomaschke Under 37 C.F.R. § 1.132 dated June 18, 2003 and submitted by Appellant as part of his Response to a final rejection filed on June 18, 2003.

Exhibit 3: "Information Communication" of Appellant entered into record by Appellant as Appendix B to Appellant's Amendment filed May 7, 2004.

Exhibit 4: Declaration/Affidavit of John E. Tomaschke Under 37 C.F.R. § 1.132 dated May 6, 2004 and submitted by Appellant as part of his Response to a final rejection filed on May 7, 2004.

B. Prior Art cited by the Examiner:

Exhibit 5: U.S. Patent No. 6,063,278 to Koo et al.; issued May 16, 2000; cited by the Examiner in the Office Action of June 19, 2002.

Exhibit 6: U.S. Patent No. 4,983,291 to Chau et al.; issued Jan. 8, 1991; cited by the Examiner in the Office Action of October 23, 2002.

C. Authorities cited in Appellant's legal analysis:

Exhibit 7: Decision in *In re Koller et al.*, 613 F.2d 819, 204 U.S.P.Q. 702 (C.C.P.A.: 1980).

Exhibit 8: Grant, HACHK'S CHEMICAL DICTIONARY (4th ed.), p. 646 (McGraw-Hill Co., 1969)

Exhibit 9: Lewis, HAWLEY'S CONDENSED CHEMICAL DICTIONARY (13th ed.), p. 1060 (John Wiley & Sons, 1997)

X: Appendix C: Related Proceedings:

There are no other proceedings related to this application or appeal.



Optimizing the Performance of Low Fouling Membranes for the World's Largest Nanofiltration Plant

by

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Abstract

The City of Boca Raton Florida is currently constructing a 40-mgd nanofiltration plant. This will be the largest plant of its kind in the world when it is commissioned in late 2003. This paper focuses on the design features of the plant and the results of extensive pilot and bench-scale tests. The pilot and bench-scale tests were conducted to find ways to reduce the severe fouling caused by the very high organic content of the raw ground water. Some interesting results from the testing are discussed including zeta potential and electrophoretic testing and side-by-side comparisons of various membranes and antiscalants.

Introduction

In order to reduce finished water color levels and enhance their ability to meet drinking water regulations for disinfection by-products (DBP), the City of Boca Raton is in the process of constructing a 40-mgd nanofiltration plant—the largest of its kind in the world.

In recent years, the City of Boca Raton, Florida, has experienced a gradual, but steady increase in the levels of color in the raw and finished water at its Glades Road Water Treatment Plant. In addition, high levels of dissolved organics in the raw water have made compliance with new, more stringent regulations for DBPs more difficult with the existing lime softening process. While the conventional lime process does remove some color, DBP precursors, and associated constituents, it is not capable of consistently and reliably meeting both DBP and color standards simultaneously. Faced with increasing customer dissatisfaction with high color levels in the finished water and stricter drinking water regulations for DBPs, the City recognized the need to pursue other treatment process alternatives to meet its drinking water quality objectives.

In order to meet these goals, the City evaluated several alternatives for process modifications, including enhanced lime softening, ozonation, and membrane softening. Due to the efficiency of the membrane process in simultaneously removing color and DBP precursors, the City selected the membrane softening process to enhance their existing treatment facilities. This technology is being used increasingly in Florida as an

alternative (or a complement) to conventional lime softening to treat relatively high colored groundwater supplies such as the City's. Nanofiltration membranes provide a way of removing over 90 percent of the total organic carbon (TOC) from the raw water therefore eliminating the majority of the precursors to DBPs. In addition, pilot testing of nanofiltration at the City produced finished water with a color of less than two color units (CU).

The Camp Dresser & McKee (CDM) project team includes CH2M Hill and laboratory research on membrane fouling has been conducted by Dr. Harvey Winters.

Early Pilot Testing

Early in the design phase of this project, it was recognized that the raw water at the Glades Road Water Treatment Plant (WTP) presented several challenges for developing a successful membrane treatment process. A membrane pilot test program was initiated to address pretreatment concerns and to develop a membrane treatment process design that would meet the City's water treatment objectives. The pilot testing revealed that cartridge filter and membrane fouling was a significant problem that needed to be addressed in the design. Fouling issues included sand and silt, oxidation of iron and hydrogen sulfide, biofouling, and dissolved organic foulants. Due to the extent of fouling encountered in the pilot testing, it was evident that the raw water quality must be improved through rehabilitation of the existing wellfield and/or installation of multimedia pressure filters. Initial fouling of the cartridge filters was so rapid that filter cartridge replacement was required approximately every seven days. To address this problem, a multimedia filter was installed upstream of the cartridge filters to lengthen their run time.

After the installation of the multimedia filter, a significant improvement was seen in the operating life of the cartridge filters; however, membrane fouling continued to be an issue. The pilot plant operating data still showed a continuous decline in membrane flux due to dissolved organic foulants. These flux decline trends in all three stages of the membrane pilot plant are shown in **Figure 1**.

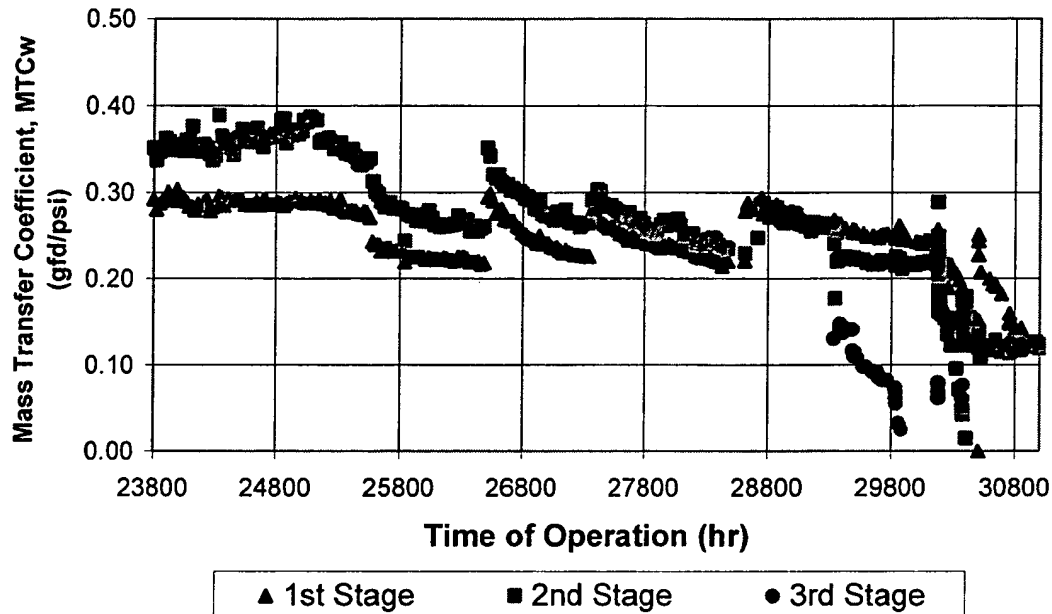
A variety of pretreatment chemical combinations and membrane models were investigated in an effort to alleviate these fouling problems. Pilot testing activities aimed at addressing this membrane fouling problem included:

- Demonstration of operation without acid addition
- Side-by-side comparisons of several membranes with respect to fouling characteristics and hardness rejection
- Side-by-side comparisons of antiscalants and dispersants to assess impacts on fouling

The results of these tests are summarized in **Figure 1**. During the operating period from 25500 to 30800 hours, four test runs using different combinations of acid and antiscalants were conducted. These tests included several brands of the most commonly used antiscalants and antifoulants in the industry. These tests were characterized by membrane fouling and rapid declines in membrane mass transfer coefficient. Fouling at

these rates would require cleaning at less than three month intervals, which was considered to be unacceptable.

FIGURE 1. MASS TRANSFER COEFFICIENT VS. TIME OF OPERATION



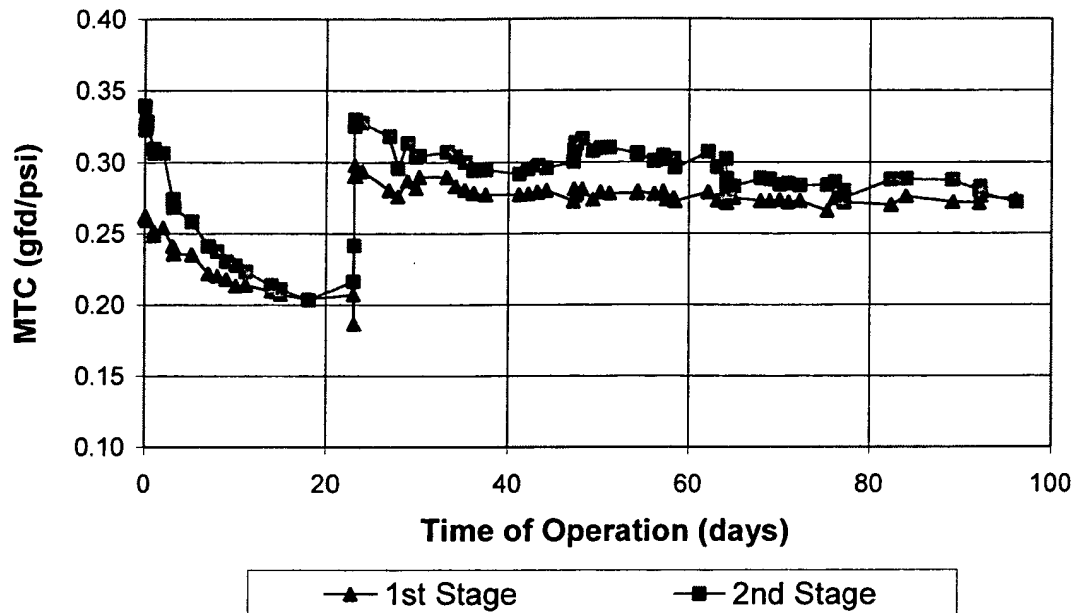
Operation without Acid and Antiscalant

After several unsuccessful test runs using some of the most widely used pretreatment chemicals in the membrane industry, several membranes were removed from the pilot test unit for autopsy analysis. The results of this testing revealed that, while substantial fractions of the foulants were consistent with naturally occurring humic acids, other components consistent with the active ingredients of the antiscalant chemicals were also observed in significant concentrations. The results of the membrane autopsies and observations from other membrane treatment plants in South Florida with high humic acid concentrations appeared to indicate that these antiscalants may actually be complexing with the naturally occurring humic acids and may be contributing to the fouling problem. To test this theory, it was decided to operate the pilot unit without either acid or antiscalant addition.

A comparison of operation with and without antiscalant addition is illustrated in **Figure 2**. The initial 20 days of operation is indicative of pilot unit performance using antiscalant with no acid addition. Operation during this period is characterized by fouling and a rapid decline in mass transfer coefficient in both stages. Testing from day 22 through the remainder of the test run was conducted without acid or antiscalant pretreatment chemicals. Although there was slightly more than a 10 percent decline in

mass transfer coefficient in the second stage, operation during this period was much more stable than in the previous test and the testing summarized in **Figure 1**.

FIGURE 2. MASS TRANSFER COEFFICIENT VS. TIME OF OPERATION



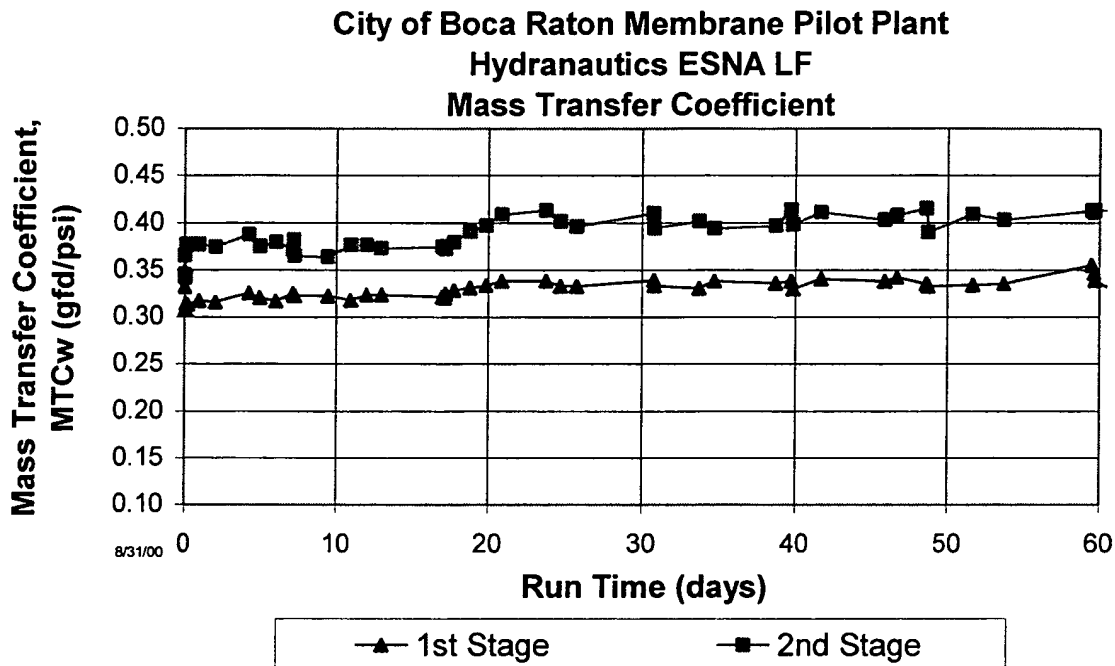
New Low Fouling Membranes

Following the initial successful trial run without acid and antiscalant, it was decided to replace the membranes, which had been installed in the pilot unit during the various pretreatment chemical trial runs, with a new set of membranes. The membranes selected for this testing were a new prototype model from Hydranautics that were developed specifically to have a reduced fouling potential for waters with high humic acid concentrations. These membrane elements were predecessors to what would later become the ESNA LF line of low fouling nanofiltration membranes. The ESNA LF membranes will use similar low fouling technology, which has already been applied to their LF series of membrane for low pressure RO applications.

The results of the first sixty days of testing with the new low fouling membranes are presented in **Figure 3**. A minor adjustment was made to the pilot unit operation between day 17 and 20; however, it can be seen that the mass transfer coefficients during this period of operation were very stable. These results represented the best performance with respect to fouling that had been achieved with the pilot testing up to that point.

The pilot test unit continued to operate with the ESNA LF membranes for approximately six to eight months without acid and antiscalant addition. Operation during this period was generally stable with respect to humic acid fouling; however, there were periods in which SDI's from the wellfield were higher than recommended for pilot unit operation.

FIGURE 3. ESNA LF MASS TRANSFER COEFFICIENT VS. TIME



This problem was compounded by a drought situation that resulted in water use restrictions. The drought lowered groundwater levels, which resulted in entrainment of air in some wells with marginal submergence over the pump impellers. In addition, the water use restrictions resulted in frequent cycling of the wellfield between watering and non-watering days. Both the lower groundwater levels and more frequent cycling of the wellfield resulted in increased air entrainment and possibly more solids production from the wells. Due to the presence of hydrogen sulfide and iron in the raw water, this air entrainment resulted in increased formation of colloidal sulfur and ferric hydroxide and unacceptable SDI readings.

The problem with high SDI's was resolved by providing a new raw water connection for the membrane pilot unit. This new raw water connection allowed the pilot unit to operate exclusively on water from the northwest wellfield as opposed to a composite blend of all wellfields. The northwest wellfield will now be the primary raw water supply for the new full-scale membrane plant; therefore, this configuration is considered to be more representative of actual full-scale plant operating conditions. After the installation of the new raw water line, SDI's were reduced to the range of 2.0 to 2.5 and allowed pilot testing to resume.

Side-by-Side Testing

Having demonstrated satisfactory operating performance without acid and antiscalant and having resolved the high SDI problem, it was decided that membranes from three major

membrane manufacturers should be tested side-by-side in the pilot test unit. The primary objectives of this testing were:

- Verify that the project water quality objectives could be achieved and specifically determine that the membranes would operate within the required applied pressure criteria and would produce water in the proper hardness range.
- Confirm that membranes from approved membrane manufacturers could perform satisfactorily without acid and antiscalant addition.
- Confirm that membranes from approved membrane manufacturers perform in accordance with their performance projection programs.

The primary permeate water quality requirements for the membrane treatment plant design with the corresponding raw water values are summarized in Table 1. As discussed

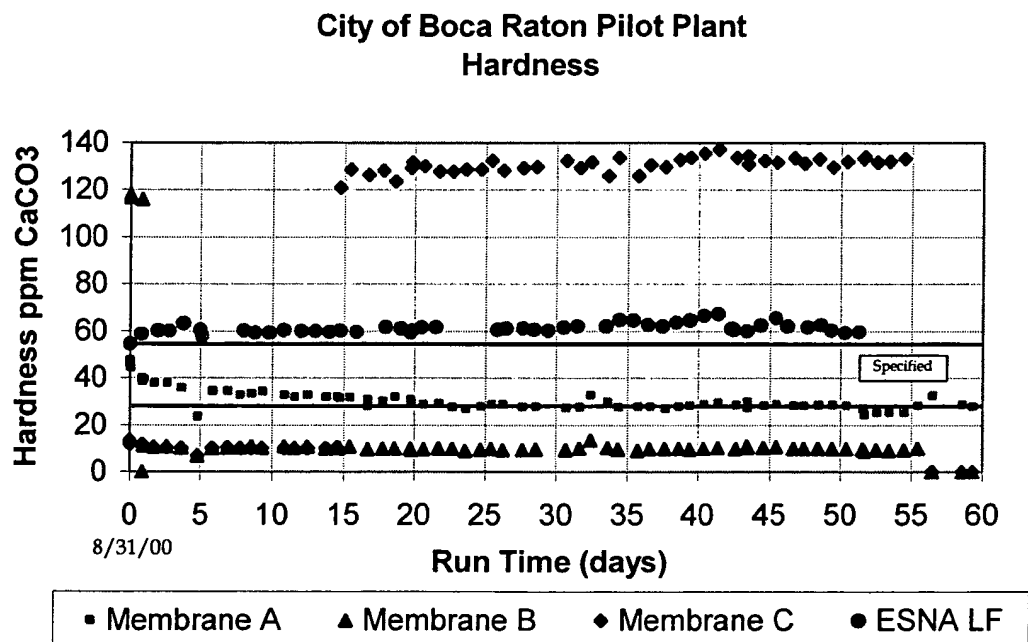
TABLE 1 – REQUIRED WATER QUALITY

Parameter	Unit	Projected Raw Water	Permeate Quality Stage 1&2
Bicarbonate*	Mg/L	265	<175
Color	CU	50	<2.0
Sum of Ions	Mg/L	450-500	<300
Total Hardness	Mg/L as CaCO ₃	250	50-80
Total Organic Carbon	Mg/L as C	12.0	<1.0
Total Trihalomethane** Formation Potential	Mg/L	0.60	<0.042
Haloacetic Acid Formation Potential***	Mg/L	0.40	<0.030

in the introduction, primary water quality parameters of concern for this project include color, hardness and disinfection byproduct formation potential. The blending rate of lime softened water with membrane softened water was selected to result in a finished water color of approximately 5.0. Since the lime softened water typically averages 12 CU, a 2:1 blending rate of membrane softened to lime softened water will result in the required finished water color. For corrosion control purposes the City prefers to maintain finished water in a hardness range between 70 and 90 mg/l as CaCO₃. Based on the selected blending ratio and typical lime softened water hardness, the permeate from the membrane softening facility should have a hardness in the range of 50 to 80 mg/l as CaCO₃ to meet this objective. Similarly permeate THM and HAA formation potential need to be within the specified ranges to satisfy the finished water quality requirements.

Three major nanofiltration membrane element manufacturers were asked to provide membrane performance projections for the full-scale membrane plant and seven of the corresponding model of 4-inch diameter membrane elements for the side-by-side testing. Based on a review of these membrane performance projections, the maximum design transmembrane pressure for this project was set at 80 psi. In general, elements from all three manufacturers were able to operate within the required transmembrane pressure.

FIGURE 4. ESNA LF PERMEATE HARDNESS VS. TIME

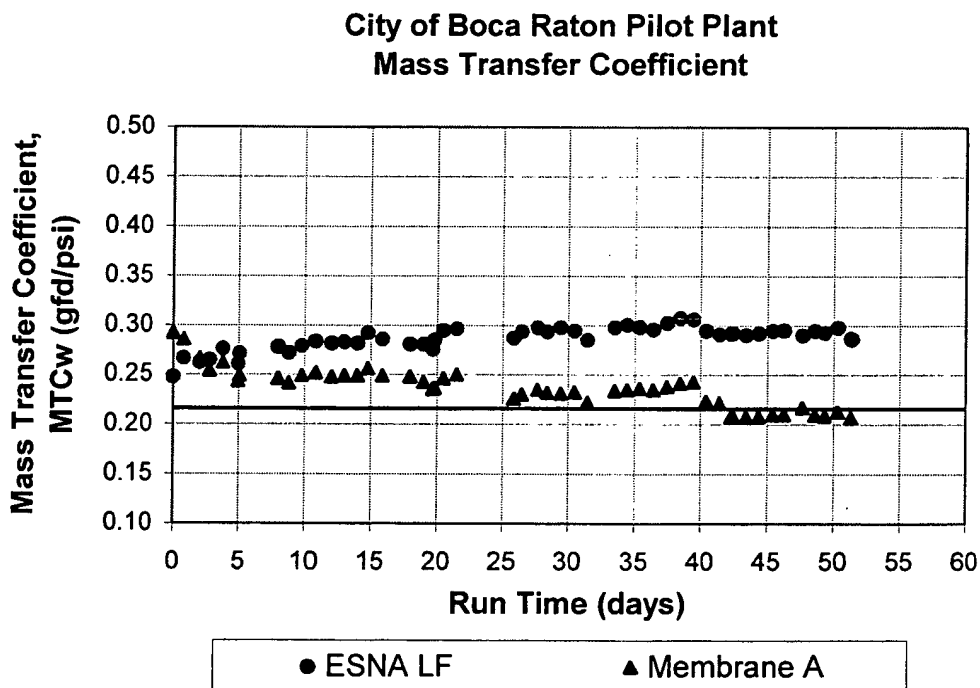


In contrast, only one of the membrane element manufacturers was able to produce permeate in the specified hardness range. A graph of permeate hardness versus time for the side-by-side testing is provided in Figure 4. This graph illustrates that the membranes initially provided by both membrane manufacturers B and C produced permeate with hardness well below the specified range. Manufacture C subsequently provided an alternate membrane element with lower hardness rejection. These new membrane elements were installed in the pilot unit on day 15 and as shown in the graph, the permeate hardness produced by this membrane was well above the desirable range. Neither manufactures B or C were able to offer hybrid designs acceptable to the City which could meet the permeate hardness target.

Only the membrane from manufacturer A produced water in the specified hardness range. Although the ESNA LF membrane was not originally included in the side-by-side testing due to price considerations, this membrane was subsequently added to the testing because of its more stable operating performance. As shown in **Figure 4** although membrane A produced permeate hardness in the proper range, the membrane exhibited more susceptibility to humic acid fouling as evidenced by decreasing hardness passage with time.

The fouling tendency of membrane A is further illustrated in the plot of mass transfer coefficient vs time presented in **Figure 5**. In this side-by-side test, the mass transfer coefficient of membrane A decreased by approximately 33 percent. In contrast, the mass transfer coefficient for ESNA LF membrane was very stable and actually exhibited a slight increase over the first few weeks of operation.

FIGURE 5. MASS TRANSFER COEFFICIENT VS. TIME

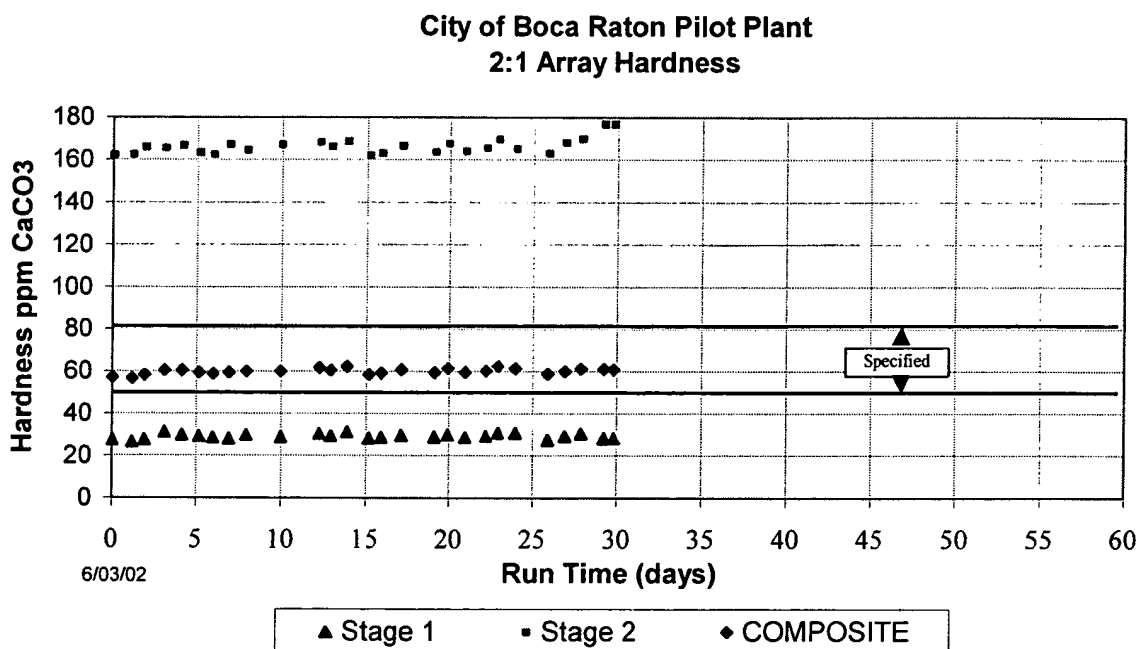


2:1 Array Testing

As shown in **Figure 5** the permeate hardness of the ESNA LF membranes which were originally tested, produced permeate with a hardness just above the desirable hardness range. Through the course of the side-by-side testing, Hydranautics exhibited greater flexibility in optimizing the performance of the ESNA LF to meet the permeate quality requirements. A series of ESNA LF membranes were produced with hardness rejections of 92, 95, and 98 percent. To demonstrate that the ESNA LF membrane could be made to meet the required permeate hardness range for this project, a 2:1 membrane array was tested with 95 percent hardness rejection ESNA LF membranes in the first stage and 92 percent hardness rejection membranes in the second stage.

The results of the 2:1 array testing are shown in **Figure 6**. The graph illustrates the permeate hardness for the first stage, second stage and composite total permeate hardness. The composite permeate hardness for the 2:1 array was approximately 60 mg/l which is close to the middle of the desirable permeate hardness range.

FIGURE 6. PERMEATE HARDNESS VS. TIME



Summary

Although numerous problems were encountered in developing the design for the largest nanofiltration plant in the world, through a comprehensive pilot testing program these problems were overcome and an optimum membrane treatment solution was developed. Initial severe colloidal fouling problems were solved through a combination of wellfield rehabilitation and installation of multimedia filtration. When traditional chemical pretreatment options failed due to unacceptably high fouling rates associated with naturally occurring organics, an unconventional approach was adopted that eliminated pretreatment chemicals and put the naturally occurring organics to work as scale controlling agents. Not only does this approach simplify operation and avoid potential problems associated with acid storage and handling, but more importantly it significantly reduces operating costs. Initial calculations indicated that this treatment facility would require one tanker truckload of acid per day at a cost of approximately \$3,000 per day. In addition to cost savings associated with eliminating acid, similar savings will also be realized due to the elimination of antiscalant.

Another revolutionary development, which evolved through this pilot test program, was the introduction of low fouling technology to nanofiltration membranes. The viability of

long-term operation without acid and antiscalant was first demonstrated using the new low fouling ESNA LF nanofiltration membranes. These membranes demonstrate much less susceptibility to organic fouling than several other conventional nanofiltration membranes in side-by-side testing.

The new low fouling membranes initially were not included in the side-by-side testing due to capital cost considerations. Hydranautics advised that, as with their low fouling LF series for brackish water treatment, there were additional manufacturing costs associated with producing the low fouling ESNA LF nanofiltration membranes. Hydranautics indicated that the additional manufacturing costs associated with producing the low fouling membrane surface would necessitate a 15 to 20 percent premium over the cost of conventional nanofiltration membranes. For this reason, there was initially a concern that the ESNA LF membrane may not be cost competitive with other nanofiltration membrane elements. However, considering that the membranes from manufacturers B and C did not meet the permeate hardness target and considering that membrane A exhibited troublesome fouling tendencies, it was decided to add the ESNA LF membrane to the side-by-side testing. The results of this testing indicated that the ESNA LF membrane offered the following advantages:

- Much more stable and reliable operation than the Membrane A.
- Lower fouling tendency than the other membranes. The lower fouling tendency should result in reduced cleaning frequency and cleaning costs.
- Lower operating pressure (10 -12 psi lower transmembrane pressure) than the Membrane A. This should result in lower operating costs.
- Better THMFP rejection than Membrane A.
- Ability to meet the permeate hardness requirements.

Most importantly the operational benefits and cost savings associated with the elimination of acid and antiscalant will more than offset the small capital cost premium for these membranes. In addition, through further pilot testing, the properties of the ESNA LF membrane were optimized to meet the treatment City's treatment objectives. The hardness rejection of these membranes was optimized to produce permeate in the City's desired hardness range. This was an important objective for the City since it reduces corrosion control concerns and post-treatment costs and it simplifies operation with respect to blending permeate with lime softened water. These benefits were achieved while maintaining the ability to meet treatment objectives for color and DBPs. In addition to setting a new precedent in terms of nanofiltration capacity, this project introduced several innovations in the approach to pretreatment and in low fouling technology for nanofiltration.

Hybrid Membrane Design

At the time that the Deerfield Membrane Treatment plant was designed, the Hydranautics ESNA1 membrane was a relatively new nanofiltration membrane available on the market. The manufacturer's specification sheet rated this nanofiltration membrane at approximately 80 percent salt rejection. The corresponding hardness rejection was on the order of 94 percent. Membrane performance projections using the manufacturer's proprietary software program indicated that a membrane system design using the ESNA1 membrane alone would not provide sufficient hardness removal.

To overcome this problem a hybrid membrane design was developed which used higher rejection ESPA1 membranes in the first stage together with the lower rejection ESNA1 membranes in the second stage. The ultra low pressure ESPA1 membranes in the first stage provided the required removal of hardness and other dissolved ions, while installation of the nanofiltration membranes in the second stage resulted in the production of a more balanced finished water and superior flux distribution in the membrane treatment process. This hybrid design took advantage of the lower power consumption associated with the ESNA1 membrane, while incorporating the higher rejection properties of the ESPA1 membrane to meet the quality objectives. Table 2 presents a performance comparison of the ESPA1 and ESNA1 membranes. As can be seen from this table, the ESNA1 membrane produces 10,000 gpd of permeate at an applied pressure of 75 psi compared to a permeate production of 12,000 gpd at an applied pressure of 150 psi for the ESPA1. The dramatically lower operating pressure for the ESNA1 membrane would translate into reduced power consumption and lower operating costs.

Table 2 also illustrates the dramatic difference in salt rejection between the two membranes. The ESPA1 membrane has a high salt rejection rate of 99 percent whereas the ESNA1 membrane has a nominal salt rejection of 80 percent. As noted above, at this lower rejection, computer projections indicated that the ESNA1 membrane alone would not be capable of achieving the permeate quality objectives for this project.

Table 2 Membrane Performance Comparison

Membrane Model	ESPA1	ESNA1
Permeate Flow	12,000	10,000
Salt Rejection (minimum):	99.0%	80.0%
Hardness Rejection	99.7%	94.0%
Configuration	Spiral Wound	Spiral Wound
Material	Polyamide	Polyamide
Nominal Membrane Area	400 ft ²	400 ft ²
Standard Test Conditions		
NaCl Solution Concentration, ppm	1500	500
Operating Pressure, psi	150	75
Operating Temperature	25	25
Recovery	15%	15%
pH	6.5 – 7.0	6.5 – 7.0

It was decided that four membrane trains producing 2.625 mgd each should be provided to produce the required permeate capacity of 10.5 mgd. A membrane array of 48:24 was selected for each 2.625 mgd train to achieve an appropriate permeate flux for the system. For the hybrid system design ESPA1 membranes would be used in the first stage with ESNA1 membranes in the second stage. This configuration provides the most balanced flux distribution for the system and avoids the need for repumping between the first and second stages.

A comparison of the performance of the membranes in each stage is provided in Table 3. This table illustrates the much higher rejection rate for the ESPA1 elements in the first stage versus the ESNA1 elements in the second stage. The ESPA1 elements were projected to produce permeate with a TDS of only 23 mg/l and a hardness of less than 10 mg/l. In comparison, permeate from the second stage was projected to have a TDS of 488 mg/l and a hardness of 155 mg/l. This would result in a combined permeate with a TDS of 164 mg/l and a hardness of 54 mg/l. This table also illustrates the well balance flux distribution between the first and second stage with the first stage operating at a flux of 13.6 gfd compared to a flux of 11.9 gfd in the second stage. Based on this design flux distribution, approximately 70 percent of the total permeate would be produced in the first stage with the second stage producing the remaining 30 percent.

Table 3 Hybrid System Stage by Stage Performance Comparison			
	Stage 1	Stage 2	Total
Capacity, gpm	1,827,816	797,184	2,625,000
No. of Vessels	48	24	72
No. of Elements/Vessel	7	7	-
Area/element	400	400	-
Flux, gfd	13.6	11.9	-
TDS, ppm	22.8	488	164.2
Total Calcium, ppm	2.8	61	20.4
Total Hardness	9.6	155	53.7
Feed Pressure	75.6	52.7	-
Recovery	59.2%	63.2%	-
Backpressure, psi	0	0	-

Recent Developments

The nanofiltration market is changing rapidly and the performance of several membrane models has changed dramatically since the time that the design was developed for this project. For example, Hydranautics has worked to improve the rejection of the ESNA1 membrane and has increased the nominal specification sheet rejection from 80 percent for

the original Deerfield Beach design to 90 percent at the present time. Rejection of greater than 94 percent has been observed from membranes used for pilot testing. Koch has made similar increases in rejection for their 8921 series of nanofiltration membranes. In addition, Dow has made changes to their nanofiltration lineup by replacing their NF70 membrane with a higher rejection NF90 membrane and a lower rejection NF200 membrane. For many membrane softening applications in South Florida, this means that a hybrid design is no longer needed to achieve the required reduction in hardness. This will allow municipalities to take full advantage of the lower operating pressure and lower power consumption of nanofiltration membranes not only in the second stage but throughout the entire membrane array.

In addition, Hydranautics is in the process of releasing a new lower fouling version of the ESNA membrane that may be commercially released as the ESNA LF (low fouling) membrane. This membrane will use similar low fouling technology, which has already been applied to their LF series of membrane for low pressure RO applications. These lower fouling membranes offer an opportunity to significantly reduce operating costs by lowering power consumption and reducing the cost of chemicals and labor associated with more frequent membrane cleaning. It has been observed that naturally occurring organics have a tendency to foul the membrane surface. As a result of this fouling, the applied pressure to the membranes must be increased to maintain the required permeate flow. This results in an increase in power consumption and the associated operating costs. In addition, more frequent membrane cleaning may be required to control membrane fouling and prevent the fouling from becoming excessive and/or irreversible. By lowering membrane fouling tendency, system reliability will be improved, and system down time and lost production associated with membrane cleaning will be reduced.

Another benefit of the ESNA LF membrane is that its hardness rejection characteristics have been tailored to provide the optimum hardness rejection for softening applications in South Florida. A number of commercially available nanofiltration membranes were tested side by side in a recent pilot test for another city with similar hardness characteristics as the City of Deerfield Beach. In that testing the ESNA LF membrane reduced hardness into the range of 85 to 95 ppm. In this hardness range the permeate is very compatible with the water produced in the lime softening process but with lower color and DBP formation potential. This hardness range is also consistent with the 80 ppm desirable hardness goal set in the AWWA study mentioned previously. The other commercially available nanofiltration membranes, which were evaluated in the test, tended to produce permeate hardness which was either far below (20 ppm) or high above (120 ppm) the optimum range. Permeate with the lower hardness values would either require a higher blending rate of permeate with raw water or lime softened water or would require stabilization with treatment chemicals such as lime or calcium chloride to increase hardness into the desirable range. In a number of applications additional blending may be problematic due to other limiting factors such as color and or disinfection byproduct formation potential. Another alternative would be to accept the lower hardness and use a corrosion inhibitor for corrosion control. The addition of a stabilization chemical and/or a corrosion inhibitor would result in an associated increase in operating cost.

Low Fouling Membrane Performance Comparison

Mechanisms which facilitate membrane fouling by organic molecules include membrane surface charge, concentration of organics, membrane flux and operating recovery. Previous investigations have demonstrated that different membrane models have different surface charges, which may make that model more susceptible to fouling by organics. Figures 2 and 3 show test results from a side by side test of the ESNA LF membranes versus another nanofiltration membrane (membrane A) and a hybrid design (Figure 3). Figure 2 illustrates that membrane A experienced a rapid decline in mass transfer coefficient from startup through the first 30 to 40 days of operation. The mass transfer coefficient for membrane A declined by approximately 30 percent while the ESNA LF membrane initially showed a slight increase in mass transfer coefficient and then exhibited very stable operation for the duration of the test. The reference line on the graph indicates the mass transfer coefficient corresponding to the maximum specified membrane operating pressure. If the mass transfer coefficient falls below this value, the maximum specified operating pressure of 80 psi (transmembrane pressure) would be exceeded.

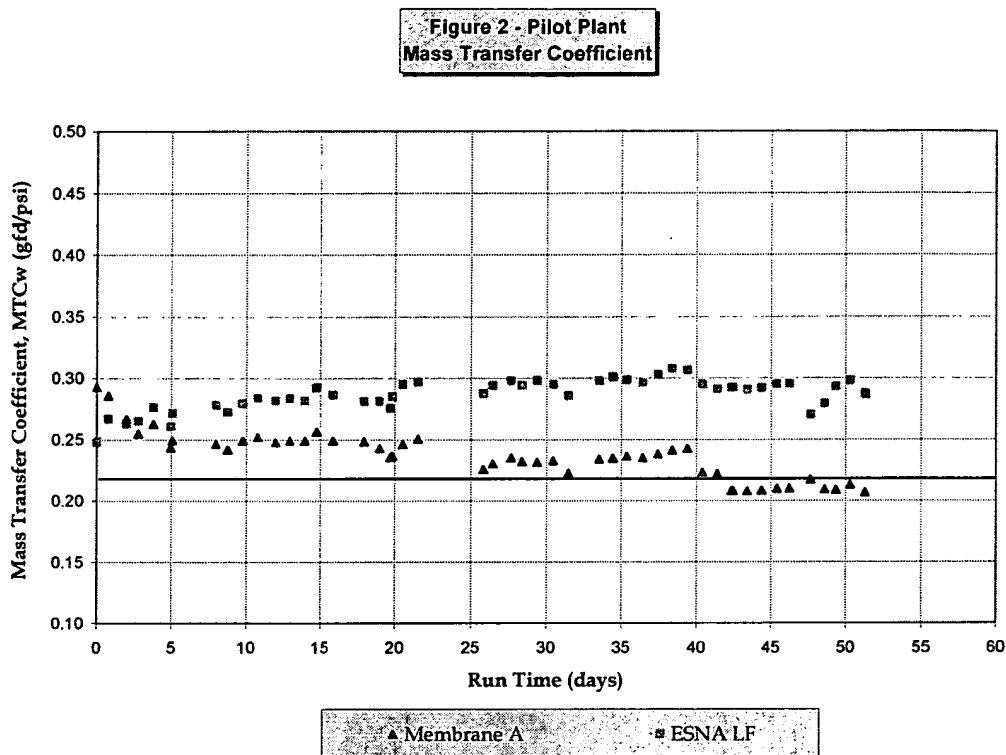


Figure 3 presents a comparison of hardness passage for an ESNA LF system with a Hybrid membrane design using an ultralow pressure Membrane B in the first stage and the nanofiltration Membrane A in the second stage. As can be seen from this graph, the hardness passage for membrane A decreased by greater than 50 percent over the first 30 days of testing. The hardness passage of the ESNA LF membrane in similar side by side testing remained relatively constant. This graph illustrates that the permeate hardness for the Hybrid membrane design would be well below the desirable range of 50 to 80 ppm which is indicated by the two reference lines on the graph. Permeate produced by the ESNA LF membrane is much closer to the desirable range and is very consistent with the 70 to 90 mg/l of hardness which is typically produced by the lime softening plant. With the low permeate hardness of the Hybrid design, additional post-treatment may be required to stabilize the finished water as discussed previously.

Figures 2 and 3 illustrate the superior operating stability of the ESNA LF membrane. The rapid decline in mass transfer coefficient and hardness passage for membrane A indicates that the performance of this membrane was dramatically affected by fouling. In addition to increased power consumption and cleaning costs associated with this fouling, there is also a concern that cleaning to restore the mass transfer coefficient and lower operating pressure may also significantly increase hardness passage to the initial values. This could result in wide swings in hardness values associated with membrane cleanings.

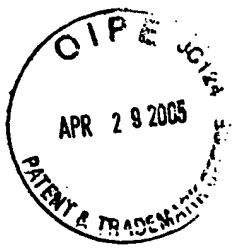
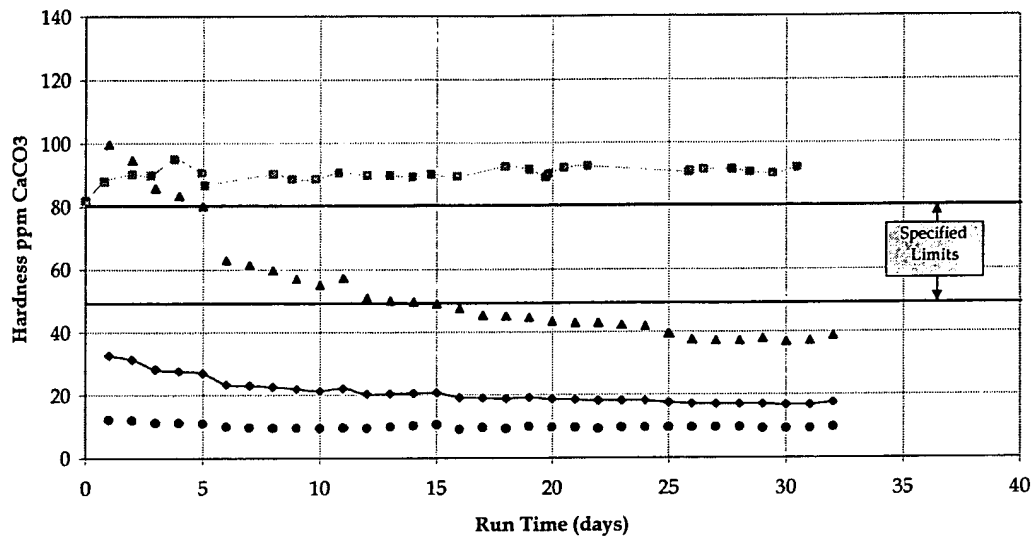
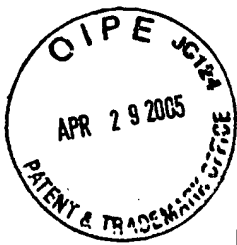


Figure 3 - Permeate Hardness
Low Fouling vs Hybrid



8/31/00

▲ STG 2 - Membrane A ● STG 1 - Membrane B ◆ Total ■ ESNA LF



PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of)	FOR: LOW PRESSURE REVERSE
)	OSMOSIS AND NANOFILTRATION
JOHN E. TOMASCHKE)	MEMBRANES AND METHOD FOR
)	THE PRODUCTION THEREOF
Serial No.: 09/724,883)	
)	Group Art Unit: 1723
Filing Date: November 28, 2000)	

DECLARATION/AFFIDAVIT OF JOHN E. TOMASCHKE
UNDER 37 C.F.R. § 1.132

I, JOHN E. TOMASCHKE, hereby declare and state that:

1. I am the inventor named in the above-identified patent application. I am Director of Membrane Development for Hydranautics, Inc., the Assignee of the present application. I have practiced in the field of reverse osmosis, nanofiltration and membrane technology for a number of years and hold scientific academic degrees consistent with such practice and with the position I now hold.
2. I am familiar with all prior proceedings in this application before the Patent Examiner, including the most recent Office Action dated March 26, 2003.
3. I have read the references cited in that Office Action, namely U.S. Patent No. 4,983,291 to Chau, Light and Chu (assigned to Allied-Signal Inc.) and U.S. Patent No. 6,083,278 to Koo and Yoon (assigned to Saehan Industries Inc.). I am familiar with the two technologies described in these patents and fully understand the disclosures of the references. I have discussed these references and the Office Action with my patent counsel and am advised that the legal basis of the Examiner's rejection of the present claims in the patent application is that, in the Examiner's opinion, if a person skilled in the art combined the teachings of the Chau and Koo patents in the manner identified by the Examiner in the Office Action, that person would consider my claimed invention to be obvious from those combined teachings.
4. In order to support my contention in the accompanying Response to Final Rejection Under 37 C.F.R. § 1.113 that the Examiner's opinion of obviousness is incorrect, I have

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conducted technical tests in which I compared examples my invention directly with equivalent examples of their inventions described by Chau and Koo in their patents. In the Table on the next page I have identified my comparative examples as "Comp." A, B, C and D . The Chau and Koo examples are clearly identified. The comparisons are of the respective formulations, processes and performances of the three inventions.

5. In the Table several abbreviations are used. These abbreviations are listed below along with their meanings.

DABCO = 1,4-diazabicyclo(2,2,2)octane

MPD = m-phenylene diamine

TMC = trimesoyl chloride

Isopar® = commercial isoparaffic solvent product from Exxon Corp.

MSA = methanesulfonic acid

PTSA = p-toluene sulfonic acid

ESPA® = commercial composite polyamide RO membrane from Hydranautics Corp., described in detail in U.S. Patent No.5,576,057.

gfd = gallons/ft²-day

'883 = membrane made according to the present invention

The tests were all conducted at 75 psig and a solution of 2000 ppm NaCl and are with respect to membranes for use in water softening.

(Table follows on next page;

Text resumes on page 4)

TABLE

Example	Membrane Substrate	Reactive Amine Formula	Crosslink Formula	Post Treatment	Drying	Flux (gfd)	Rejection (%)
Koo, Exp. 1	polysulfone	MPD, DABCO, MSA	TMC in ISOPAR®		90°C for 3.5 mins	4.3	98.0
Comp. A	same	MPD, MSA	same		same	2.4	97.7
Comp. B	same	ESPA®	ESPA®		similar	13.0	98.2
Comp. C	ESPA®			DABCO, MSA	90°C for 2 mins	14.0	98.5
Chau, Exp. 5	same*			PTSA	170°C for 2 mins	6.6	95.2
Comp. D	same*			PTSA	90°C for 2 mins	13.8	98.2
'883	same*			MSA	same	28.4	87.4

* Chau's membrane substrates and those of the present invention's compositions are essentially equivalent to the ESPA® product with respect to properties relevant to comparison as to post treatment technology. Therefore to expedite the collection of these data, Applicant has used the commercial ESPA® product as the functional equivalent for these membrane substrates. Such use does not alter the results of the tests or the validity of the comparisons for patent purposes in any manner.

6. From the data in the Table, the following conclusions can be drawn.
- a. The Koo formulation system, which does not use any post-treatment step, results in an RO membrane which has a low flux and high salt rejection (Koo, Exp. 1). This type of membrane would be unsuitable for nanofiltration and water softening. Removing the DABCO tertiary amine from Koo's amine salt formulation results in a membrane with even lower flux and continued high rejection (Comp. A)
 - b. Substituting a system equivalent to Hydranautics' commercial ESPA® product but without post treatment improves the flux significantly (Comp. B). Using the commercial ESPA® product as designed with post treatment provides even better flux (Comp. C).
 - c. Chau's system, which has post treatment but with a sulfonic acid outside the carbon range of C₁-C₆, has a poor flux rate and high rejection, not much better than those of Koo (Chau, Exp. 5). The flux rates of both Koo's and Chau's inventions are well below the minimum required for water softening applications. Reducing the amount of drying for the Chau system results in a product with somewhat better flux, but the rejection rate is increased back to the level of Koo (Comp. D).
 - d. The present invention, however, as shown in the last line of the Table ('883), which has post treating with a sulfonic acid having a carbon number in the claimed range of C₁-C₆, has greatly improved flux (4 times higher than Chau, 7 times higher than Koo, and 2 times higher than Comp. D) along with markedly reduced rejection over all of the other systems.
7. I believe that it would be apparent to one skilled in the art from these data that neither Koo nor Chau would lead one to the present invention. Variations to focus on improvements in Koo or Chau individually (Comps. A, B and D) do not provide more than limited improvement. The commercial ESPA® product of Applicant's assignee can be considered to be an example of a system somewhat like what the Examiner is relying on in his rejection, i.e., using a Koo-like system but with a Chau-like post-treatment (Comp. C), but it will be seen that the results still are well below the properties obtained by the

Ser. No. 09/694,431

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product of this invention ('883).

8. I have been advised by patent counsel that comparative testing submitted in patent applications is required to compare the claimed invention directly with the art which is the closest to the basis of the Examiner's rejection. I believe that these data have been obtained in a manner fully in compliance with that requirement. As I have stated above, I am aware that the Examiner has cited Chau and Koo in combination in the Office Action. It is not technically possible to make usable membranes for testing by combining components from Koo into the process of Chau as the Examiner proposes in the Office Action. Therefore the closest that I or any person skilled in the art can come is to test the claimed invention against Chau and Koo separately and to examine products (such as the ESPA® product) which to some extent parallel the speculative system proposed by the Examiner, both of which I have done for this comparison.

9. This ends my statement.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 6/18/03 Signed: John E. Tomaschke
John E. Tomaschke

(Attorney Docket No. 7703-PA02)

APPENDIX B



PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of)	
)	FOR: LOW PRESSURE REVERSE
TOMASCHKE, John)	OSMOSIS AND NANOFILTRA-
)	TION MEMBRANES AND
Serial No.: 09/724,883)	METHOD FOR PRODUCTION
)	THEREOF
Filed: November 28, 2003)	
)	Group Art Unit: 1723

INFORMAL COMMUNICATION

Examiner Krishnan S. Menon
via Facsimile 703-746-8788

Attention: Examiner Menon

Dear Examiner Menon:

As we discussed by telephone, below please find some calculations and issues relevant to the prosecution of this case. When you have had an opportunity to discuss this information with your supervisor, please contact me by telephone or e-mail so that we may discuss the case. I hope that we will be able to resolve some of the outstanding issues in the case so that the prosecution may close with the issuance of a notice of allowance.

Bridging pages 3-4 of the office action mailed on July 11, 2003, you that upon normalizing the data of Chau to allow for comparison to the conditions in claim 15, the membrane of Chau would meet the requirements of the claim. The Applicants submit that an error was made in the calculation by the Examiner and that essentially none of the membranes taught by Chau would have sufficient flux

or salt rejection as required by claim 15. The Applicants concur that the reduction of the salt concentration in the instant invention reduces osmotic pressure. However, the instant invention also requires a substantially lower operating pressure, 75 psi as compared to 220 psi which substantially reduces the net driving pressure, decreasing flux.

Below please find calculations performed to normalize the data to the limitations of claim 15 in the instant application. An explanation of the calculations made is presented.

The following equations are used in the comparison of flux at different salt concentrations and operating pressures.

$$\text{Net Driving Pressure (NDP)} = \text{Operating Pressure} - \text{Osmotic Pressure}$$

$$\text{Salt passage} = 100 - \% \text{ rejection}$$

$$\text{Osmotic pressure} = \text{salt concentration in ppm} \times 0.0115$$

In the PTSA treated membrane of Chau in Example 5, Table 4:

$$\begin{aligned} \text{Osmotic pressure} &= 2000 \times 0.0115 \\ &= 23 \text{ psi} \end{aligned}$$

$$\text{Operating pressure} = 220 \text{ psi}$$

$$\begin{aligned} \text{Net Driving Pressure} &= 220 \text{ psi} - 23 \text{ psi} \\ &= 197 \text{ psi} \end{aligned}$$

$$\begin{aligned} \text{In Claim 15 Osmotic pressure} &= 500 \times 0.0115 \\ &= 5.75 \text{ psi} \end{aligned}$$

$$\text{Operating pressure} = 75 \text{ psi}$$

$$\begin{aligned} \text{Net Driving Pressure} &= 75 \text{ psi} - 5.75 \text{ psi} \\ &= 69.25 \text{ psi} \end{aligned}$$

$$\begin{aligned}\text{The NDP factor} &= \text{NDP}_{\text{Chau}} / \text{NDP}_{\text{Claim 1}} \\ &= 197/69.25 \\ &= 2.845 \times\end{aligned}$$

Therefore, the flux of the PTSA treated membrane of Chau under the conditions of claim 15 is:

$$38.6/2.845 = 13.57 \text{ gfd}$$

This is less than the 15 gfd as required in claim 1.

The salt passage would also be altered by the same NPD factor. Therefore, under the conditions of claim 15, the salt passage of Chau would be:

$$1.8 \times 2.845 = 5.12; \quad 100 - 5.12 = 94.88\% \text{ salt rejection.}$$

All of the testing of Chau was performed under a single set of conditions. The Applicants have calculated the flux for each of the membrane conditions and found that only three of the membranes of Chau have sufficiently high flux to meet the requirements of claim 1 in regard to both flux and salt rejection. They are membranes D-2, D-3 and D-4 in Table 2. The membranes have fluxes between 16.0 gfd and 23.44 gfd and salt rejections between 94.0% and 83.5%. Although these membranes meet the working specifications of Claim 1, they are prepared by treating the membrane with 2-10% citric acid. It is of note that the membranes prepared by the method of claim 15 have higher salt rejections at comparable fluxes. Therefore, the rejection of claim 1 under 35 U.S.C. 102(b) is traversed.

The Applicants further state that in view of the large number of compounds tested by Chau that did not produce membranes that meet the specifications of claim 15, it would not have been obvious to one skilled in the art to test the specific class of compounds claimed, C₁-C₆ sulfonic acids. Therefore, claim 15 is


not obvious in view of Chau and a rejection under 35 U.S.C. 103 is not appropriate.

CONCLUSIONS

The Applicants thank the Examiner for his time and consideration and look forward to a formal telephonic interview.

Respectfully submitted,

Dated: Oct 15, 2003

By: 
Colleen J. McKiernan
Attorney for Applicant
Registration No. 48,570

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Docket No.: 7703-PA02



PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application of)	
John E. TOMASCHKE)	FOR: LOW PRESSURE REVERSE
)	OSMOSIS AND
Serial No.: 09/724,883)	NANOFILTRATION
)	MEMBRANES AND METHOD
Filed: November 28, 2000)	FOR PRODUCTION THEREOF
)	
)	Group Art Unit: 1723

DECLARATION OF JOHN TOMASCHKE UNDER 37 C.F.R. §1.132

Commissioner for Patents
Washington, D.C. 20231

Attention: Examiner Krishnan Menon

I, JOHN E. TOMASCHKE, do hereby affirm and say that:

1. I am an inventor of the above named application.
2. The ESNA LF membranes discussed the references listed in the attached Appendices D, E and F are membranes made by Hydranautics, Inc., using the method of the invention.
3. The first order of an ESNA LF membrane for testing was received by Hydranautics, Inc. on January 19, 2000. The element was shipped to the individuals performing the testing for the Boca Raton plant on January 28, 2000. To the best of my ability, this is the first case of public use that I have been able to identify.

4. The ESNA LF membranes were first offered for sale in a letter of intent to the Deerfield Beach West Water Treatment Plant dated June 25, 2002.
5. The ESNA LF membranes were offered for sale to the City of Boca Raton for use in their water treatment facility that was the largest plant of its kind at the date of commission late in 2003. The facility requires about 7690 membrane elements per year to purify about 40 million gallons per day. This is substantially more than most water treatment facilities at that time that require about 192-960 membrane elements per year to purify about 1 to 5 million gallons per day.
6. Multiple membranes provided by various manufacturers were tested for use in the Boca Raton plant. One of the membranes tested was manufactured by Koch Membrane Systems, Inc., the owner of the Chau US Patent No. 4,893,291. The chain of title is attached as Appendix B. The membrane was found to be unsuitable for use in the Boca Raton plant.
7. No membranes were submitted by Saehan, the owner of the Koo US Patent 6,063,278. The chain of title is attached as Appendix G.

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like

so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 5/6/04

Signed: John Tomarchi

FULL TEXT OF CASES (USPQ FIRST SERIES)In re Koller, Hartl, and Kirchner, 204 USPQ 702 (CCPA 1980)

In re Koller, Hartl, and Kirchner, 204 USPQ 702 (CCPA 1980)

In re Koller, Hartl, and Kirchner**(CCPA)****204 USPQ 702****Decided Jan. 24, 1980****No. 79-589****U.S. Court of Customs and Patent Appeals****Headnotes****PATENTS****1. Construction of specification and claims -- Broad or narrow -- In general (§ 22.101)****Construction of specification and claims -- By specification and drawings -- In general** (§ 22.251)**Specification -- Sufficiency of disclosure** (§ 62.7)

It does not follow from fact that disclosure of specifics adds to understanding one skilled in art would glean from generic term that such added disclosure limits its meaning; indeed, neither listing of representative compounds nor example is always necessary in completely describing generic class; presence of specific examples in specification that provides statutory description via generic expression that is understandable cannot, in ex parte practice, limit that expression; claims are given broadest reasonable interpretation, consistent with specification, during prosecution of application; however, claims in issued patent may sometimes be interpreted by court in light of specification so as to protect only that phase of claimed invention that constitutes patentable subject matter and thus do justice and equity between parties.

2. Applications for patent -- Continuing (§ 15.3)**Specification -- Claims as disclosure** (§ 62.3)**Specification -- Sufficiency of disclosure** (§ 62.7)

Compliance with Section 112, paragraph one, is judged as of date application is filed; original claims constitute their own description; later added claims of similar scope and wording are described thereby.

3. Construction of specification and claims -- By prior art (§ 22.20)**Patentability -- Evidence of -- State of art** (§ 51.467)

Specification -- Sufficiency of disclosure (§ 62.7)

Later-issued patents and publications may, under certain narrow circumstances, be used to show state of art existing on date

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of application in question; these circumstances, are, for example, where later publication evidenced that as of application's filing date undue experimentation would have been required, parameter absent from claims was or was not critical, statement in specification was inaccurate, invention was inoperative or lacked utility, characteristics of prior art products were known, or claim was indefinite; however, use of later existing state of art is not permitted in determining enablement under 35 U.S.C. 112; Patent Office can rely on later-issued patents and publications to construe claim language only if showing is made that that claim language is "language of the present art" as of filing date of application in question.

4. Patentability -- Anticipation -- In general (§ 51.201)**Patentability -- Anticipation -- Carrying date back of references** (§ 51.203)

Decision of Board of Appeals affirming rejection under Section 102 is reversed where application's filing date is prior to effective date of any of cited references.

Particular patents -- Isohumulone

Koller, Hartl, and Kirchner, Method of Isomerizing Humulone to Isohumulone by Catalytic Acceleration with Metal Salts, rejection of claims 1-3 and 5-14 reversed.

Case History and Disposition:

Appeal from Patent and Trademark Office Board of Appeals.

Application for patent of Horst Koller, Alfons E. Hartl, and Gerhard Kirchner, Serial No. 582,774, filed June 2, 1975. From decision rejecting claims 1-3 and 5-14, applicants appeal. Reversed.

Attorneys:

Roger L. Browdy, Washington, D.C., for appellants.

Joseph F. Nakamura (Harry I. Moatz, of counsel) for Commissioner of Patents and Trademarks.

Judge:

Before Markey, Chief Judge, Rich, Baldwin, and Miller, Associate Judges, and Newman, Judge. *

Opinion Text**Opinion By:**

Baldwin, Judge.

This is an appeal from the decision of the United States Patent and Trademark Office (PTO) Board of Appeals (board) affirming the rejection of claims 1-3 and 5-14 in appellants' application serial No. 582,774, filed June 2, 1975, entitled "Method of Isomerizing Humulone to Isohumulone by Catalytic Acceleration with Metal Salts." ¹The claims are

rejected as anticipated under 35 USC 102. We reverse.

The Invention

Appellants claim a method for producing isohumulone from humulone. Isohumulone is a chemical said to be useful as a tuberculosis retardant. It is also the essential part of "hops-bitters" used in the brewing of beer.

The claimed invention involves the steps of forming a liquid mixture of the reactant humulone, a sufficient amount of certain metal salts (the cation of which is a member of Group IIa or IIb, or the Iron group of the periodic table, or cerium and manganese) said to be catalytic or complex-forming, and a "liquid medium." The desired isomerization takes place at an "elevated temperature," desirably at the boiling point of the system, and at a pH preferably between 4 and 9.

Claims 1 and 13 are illustrative of the invention:

1. A method for preparing isohumulone products, comprising:

providing a mixture of (1) a salt productive in an aqueous medium of an anion and a cation, said anion being inert to the starting material and inert to the isohumulone products under the operating conditions of the present method, and said cation being an element selected from the group consisting of Group IIa or IIb elements, Fe group elements, cerium and manganese, (2) humulone or a humulone-containing material, and (3) a liquid medium, inert to the starting materials and to the isohumulone produced under the operating conditions of the present method, said liquid medium being one in which said salt dissociates to form said anion and said cation and said humulone or humulone-containing material dissociates to form humulate anion; and

isomerizing at an elevated temperature, at a pH below 9 and greater than that pH

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at which humulone forms humulate anion in solution;

wherein said salt is present in an amount sufficient to accelerate said isomerization.

13. A method of preparing isohumulone products, comprising:

combining (1) humulone or humulone-containing material, (2) a liquid medium inert to the reaction materials, in which both humulone and the salt to be used are soluble, and (3) a salt productive in said liquid medium of an elemental cation of an element selected from the group consisting of Group IIa or IIb elements, Fe group elements, cerium and manganese in an amount sufficient to accelerate isomerization; and

isomerizing at an elevate[sic] temperature, at a pH lower than about 9 and above 4.

The Rejection

The examiner rejected all of the claims under 35 USC 102 as unpatentable in view of Worden, U.S. Patent No. 3,923,897, filed April 2, 1973. Worden discloses a similar process but in the main was utilized for its disclosure of Todd (Belgian patent No. 782,900 or U.S. Patent No. 4,002,683). Todd is admitted by all parties to be anticipatory of the claims (if the grandparent lacks §112 description) especially in view of its discussion of the British equivalent to the grandparent application of this case.

Background

The ultimate issue in this case is whether or not the grandparent application provides a written description of the claimed invention in the manner required by 35 USC 112. If it does, the decision of the board with regard to §102 is improper since the filing date of the grandparent is prior to the effective date of any of the cited references.

The bone of contention between the parties here is the meaning to be assigned the term "liquid medium." The basic argument stated by the PTO is that the term varies in breadth from the grandparent to this case. The appellant maintains that the term is a broad term throughout.

The examiner, in his Answer, specifically discussed the term and his view of the etymology of the term:

The grandparent application discussed the process throughout the disclosure referring to "liquid medium", "solvent", "dispersing agent", etc. However, the specific disclosure with respect to the solvent system used discloses, "As a reaction medium one can in principle use water to which, however, a water-miscible organic solvent is added as solubilizer, since humulon is practically insoluble in water. As mentioned above, water-ethanol mixtures are preferred." None of the examples in the grandparent application use other than aqueous systems consisting of water or water in addition to a watermiscible solvent such as ethanol. It is only with the filing of the continuation-in-part application, the parent of this application, now U.S. Patent No. 3,952,061, that appellants added examples drawn to the use of water-immiscible solvent systems. Appellants recognized the fact that there was no support in the original application to claim the use of a water-immiscible system which they are now trying to recoup in view of the Worden patent disclosure. It is interesting to note that in the parent application, appellants cancelled all claims directed to the use of water-immiscible solvent systems in view of a rejection over their [sic] own priority documents which had become available as prior art, * * * Up until Worden published his invention, no one had ever isomerized the alpha-acids of hops resins to iso-alpha acids in non-aqueous systems. It was Worden that[sic] discovered the great processing benefits that accrued to the use of such a system. The use of aqueous systems in place of non-aqueous systems prior to Worden was unknown. Therefore, appellants cannot allege that the use of non-aqueous systems was implied in their original disclosure since it was not specifically used, disclosed or known in the art at that time. Appellants readily admit that they added new matter in the continuation-in-part application but are attempting to imply that it was there all the time. If it were present in the parent (grandparent of this application), the new matter would not have been needed nor the additional added examples. Appellants[sic] position is believed to be in error since (1) there is no specific disclosure of the use of non-aqueous systems, the contrary being true, and (2) it cannot be inferred that the use of non-aqueous systems would have [been] readily apparent to one skilled in the art prior to Worden since the use of non-aqueous systems was the invention of Worden and not known in the art at that time.

The board affirmed the rejection and in doing so stated:

The Examiner holds that the grandparent application does not contain a description of water-immiscible solvents

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as the liquid medium in which the isomerization is effected.

The appellants argue that the term "liquid medium" set forth in the grandparent application broadly covers all solvents in which the isomerization is effected and hence encompasses the water-immiscible solvents shown by Worden.

We have carefully considered all of the arguments but we are not convinced of reversible error in the Examiner's rejection. The instant application is not entitled to the benefit of the filing date of the grandparent application since it fails to satisfy the description requirements of 35 USC 112 with respect to the disclosure of water-immiscible solvents. Although the grandparent application employs the term "liquid medium" for the isomerization of humulone, we do not find that it clearly and unequivocally describes the use of water-immiscible solvents. We find no delineation nor express description of any water-immiscible solvent either generally or specifically to serve as support for the present claims which encompass such solvent.

The use of aqueous or aqueous-alkanol media at 50°C or above, as disclosed in the earlier application, does not serve as basis for liquid medium such as methylene chloride indicated by Todd (cited by appellants) as suitable at ambient temperature. We find nothing in the grandparent to suggest the use of water-immiscible solvents or that isomerization can be effected below 50°C. The concept of isomerization taking place in the organic water-

immiscible phase is nowhere suggested, taught or described by appellants prior to the filing of their parent application on May 30, 1973. Nor, as pointed out by the Examiner, would it have been obvious to employ such solvents at that time.

Considering the disclosure as a whole, one of ordinary skill in the art at the time of the filing of the grandparent application, would have been taught how to isomerize humulone by addition of polyvalent metal salts in aqueous or aqueous alkanolic media. The broad recitation "liquid medium" would have been construed by one skilled in the art from the disclosure as consisting of water or water to which a miscible organic solvent is added. The term as now interpreted by appellants is broader than that disclosed in the grandparent application.

The subsequent discovery by Todd, referred to in Worden, of employing water-immiscible solvents for the isomerization step merely confirms the unpredictable and unobvious nature of the liquid medium and as such, requires a clearer disclosure thereof than that which appellants provide in the grandparent. The finding by Todd, for which a patent was obtained, that water-immiscible solvents could be used for isomerization below 50°C cannot nunc pro tunc be considered as part of appellants' invention as of the time of the grandparent application which is completely devoid of any concept of effecting the isomerization at low temperature, that is, below 50°C.

We do not find the issue here to be a question of enablement but to be more properly one of description in the grandparent application. The fact that the recitation "liquid medium" might include water-immiscible solvents is not sufficient indication to one of ordinary skill at that time that such medium was part of appellants' invention. In *re Winkhaus et al.*, 527 F.2d 637, 188 USPQ 129 (CCPA 1975). On the contrary, a fair reading of the grandparent disclosure would have led one to conclude that the isomerization in water and water-miscible media was appellants' contribution. Accordingly, it is our view that appellants are not entitled to the benefit of their grandparent application and the rejection of the claims under 35 USC 102 over Worden is sustained.

Opinion

[1] The solicitor argues that the recitation of the broad term "liquid medium" in the body and claims of the grandparent specification is merely "fortuitous" ²and must be understood to mean only the narrower term "water-miscible solvent." The reasoning supplied by the PTO is that disclosed narrow classes, examples and enumerated compounds must be considered in fleshing out; giving meaning to, and therefore limiting a generic expression. Certainly, the disclosure of specifics *adds* to the understanding one skilled in the art would glean from a generic term, but it does not follow that such added disclosure *limits* the meaning thereof. See *In re Fuetterer*, 50

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CCPA 1453, 319 F.2d 259, 138 USPQ 217 (1963). Indeed, neither a listing of representative compounds nor an example is always necessary in completely describing a generic class. See *In re Robins*, 57 CCPA 1321, 429 F.2d 452, 166 USPQ 552 (1970). If a specification provides a statutory description via a generic expression which is understandable, the presence of specific examples cannot, in *ex parte* practice, ³be said to limit that expression.

[2] Compliance with §112, paragraph one, is to be judged as of the date the application is filed. In *re Hogan*, 559 F.2d 595, 194 USPQ 527 (CCPA 1977); In *re Glass*, 492 F.2d 1228, 181 USPQ 31 (CCPA 1974). A comparison of the claims in this case, *supra*, with the original claims in the grandparent ⁴reveals not only that the term "liquid medium" is found in both places, but also that the two sets of claims are similar in wording. In *In re Gardner*, 475 F.2d 1389, 177 USPQ 396 (CCPA 1973), this court noted that original claims constitute their own description. Later added claims of similar scope and wording are described thereby.

Nevertheless, the PTO argues that Todd is evidence that, as of the filing date of the grandparent, one having ordinary skill in the art would not have recognized the inclusion of water-immiscible solvents within the scope of the term "liquid medium." Todd does disclose, at a subsequent time, that water-immiscible solvents are useful in the process.

[3] We recognize that, under certain narrow circumstances, later-issued patents and publications may be used to show the state of the art existing on the date of the application in question. ⁵However, the circumstances here do not fit any

exception to the general rule that language in a specification is to be understood for what it meant to one having ordinary skill in the art at the time the application was filed. In re Glass, *supra*; In re Argoudelis, 58 CCPA 769, 434 F.2d 1390, 168 USPQ 99 (1970). Todd's discussion of the invention found in the grandparent was based on his reading of the "British Patent Specification published 16 July 1969." Consequently, whatever Todd may have said concerning the meaning of the phrase "liquid medium" must be understood to have occurred at some time after July 1969 -- a time clearly subsequent to the filing date of the grandparent in April of 1968.

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This case parallels In re Hogan, *supra*. Hogan filed an application in 1953 (to which the application there in issue was related under 35 USC 120) disclosing a solid polymer produced from 4-methyl-1-pentene. The only method disclosed entailed the production of a *crystalline* polymer. During 1962 another applicant disclosed that an *amorphous* form of the polymer could be produced. The PTO challenged the scope of enablement in that 1953 specification in asserting, based on the subsequent 1962 disclosure, that the generic disclosure did not encompass amorphous polymers but only crystalline ones.

We remanded that case and stated:

Though we do not reach the point on this appeal, we note appellants' argument that their invention is of "pioneer" status. The record reflects no citation of prior art disclosing a solid polymer of 4-methyl-1-pentene, which may suggest that appellants at least broke new ground in a broad sense. * * * As pioneers, if such they be, they would deserve broad claims to the broad concept. What were once referred to as "basic inventions" have led to "basic patents," which amounted to real incentives, not only to invention and its disclosure, but to its prompt, early disclosure. If later states of the art could be employed as a basis for rejection under 35 USC 112, the opportunity for obtaining a basic patent upon early disclosure of pioneer inventions would be abolished. [Supra at 606, 194 USPQ at 537].

In Hogan, an analysis using later-filed references to determine the scope of enablement was found to be impermissible. Similarly, it cannot be allowed when, as here, the description requirement is in issue.

[4] In sum, the phrase "liquid medium" in the present claims is described in the grandparent case in the manner required by 35 USC 112, paragraph one. Consequently, the decision of the board of appeals affirming the rejection under §102 is *reversed*.

Reversed.

Footnotes

Footnote 1. This case is a continuation of application serial No. 356,785, filed May 3, 1973, (now U.S. Patent No. 3,952,061, hereinafter "the parent") which in turn is a continuation-in-part of serial No. 725,458, filed April 30, 1968, now abandoned (hereinafter "the grandparent").

Footnote 2. It is apparent that appellants specifically placed the broad phrase in the grandparent specification and supplemented it by a narrower phrase and certain examples. Why the deliberate act of the inventors in placing two phrases of varying scope in the document is "fortuitous" is manifestly unclear.

Footnote 3. Claims are given the broadest reasonable interpretation, consistent with the specification, during prosecution of an application. However, claims in an issued patent may sometimes be interpreted by a court "in light of the specification" so as "to protect only that phase of the claimed invention that constitutes patentable subject matter and thus do justice and equity between the parties." In re Prater, 56 CCPA 1381, 1395, 415 F.2d 1393, 1404, 162 USPQ 541, 550 (1969).

Footnote 4. The grandparent's original claims 1 and 2 provide:

1. A method of preparing isohumulon by isomerization of humulon or humulon-containing material at elevated temperatures in a liquid medium, characterized by the fact that the isomerization catalyzes by addition of at least one salt of a bivalent metal or of cerium.
2. A method according to claim 1, characterized by the fact that as catalyst there are used salts of elements of groups IIA or IIB of the periodic system of the elements, of the iron group of the periodic system of elements, or of cerium or manganese.

Footnote 5. These circumstances were listed in *In re Hogan*, supra at 605 n.17, 194 USPQ at 537 n.17:

Where, for example, a later publication evidenced that, *as of an application's filing date*, undue experimentation would have been required, *In re Corneil*, 52 CCPA 1718, 1724, 347 F.2d 563, 568, 145 USPQ 702, 705 (1965), or that a parameter absent from the claims was or was not critical, *In re Rainer*, 49 CCPA 1243, 1246 n.3, 305 F.2d 505, 507 n.3, 134 USPQ 343, 345 n.3 (1962), or that a statement in the specification was inaccurate, *In re Marzocchi*, 58 CCPA 1069, 1073 n.4, 439 F.2d 220, 223 n.4, 169 USPQ 367, 370 n.4 (1971), or that the invention was inoperative or lacked utility, *In re Langer*, 503 F.2d 1380, 1391, 183 USPQ 288, 297 (CCPA 1974), or that a claim was indefinite, *In re Glass*, supra, 492 F.2d at 1232 n.6, 181 USPQ at 34 n.6, or that characteristics of prior art products were known, *In re Wilson*, 50 CCPA 773, 311 F.2d 266, 135 USPQ 442 (1962). Whatever may have been said enroute to decision in these cases, the fact situation in none of them established a precedent for permitting use of a later existing state of the art in determining enablement under 35 USC 112. [Emphasis in original.]

One additional comment was made in *In re Voss*, 557 F.2d 812, 818 n.15, 194 USPQ 267, 272 n.15 (CCPA 1977):

We note the dictum in footnote 6 of *Glass* * * * [*In re Glass*, 492 F.2d 1228, 1232, 181 USPQ 31, 34 (CCPA 1974)] indicating that later-issued patents or publications may be used to construe claim language. However, it is clear from the quotation from *In re Fisher*, 57 CCPA 1099, 1106, 427 F.2d 833, 838, 166 USPQ 18, 23 (1970), set forth in the footnote that the PTO can rely on such later-issued patents and publications only if a showing is made that such claim language is the "language of the present art" as of the filing date of the application in question. The PTO has made no such showing in this case.

Footnote * The Honorable Bernard Newman, United States Customs Court, sitting by designation.

- End of Case -

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HACKH'S CHEMICAL DICTIONARY

[American and British Usage]

*Containing the Words Generally Used in Chemistry,
and Many of the Terms Used in the Related
Sciences of Physics, Astrophysics, Mineralogy,
Pharmacy, Agriculture, Biology,
Medicine, Engineering, etc.*

Based on Recent Chemical Literature

FOURTH EDITION

Completely Revised and Edited by

JULIUS GRANT

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- sulfine.** Sulfonium compound. An organic compound of the type R_3SX . See *sulfonium*.
- sulfinic acid.** $R-SO_2OH$; as, ethanesulfonic acid*, $EtSO_2H$. In derivatives it becomes the prefix sulfino-.*
- sulfimid.** Saccharin.
- sulfino*.** (1) The radical $-SO_2H$. Cf. *sulfonic acid*. (2) Prefix denoting a sulfonic functional group.
- sulfinoxide.** R_3S-OH . An organic compound.
- sulfonpyrazone.** $C_{23}H_{20}O_3N_2S = 404.5$. Bitter, white powder, insoluble in water, m.133; a uricosuric (B.P.).
- sulfanyl.** Thionyl.
- sulfon.** Sulfide ion: S^{2-} .
- sulfosoxazole.** $C_{11}H_{13}O_3N_2S = 267.32$. White crystals, m.193, insoluble in water; an antibacterial (U.S.P.).
- sulfite.** M_2SO_3 . A salt of sulfurous acid. **acid-** $MHSO_3$. **bi-, hydrogen-** Acids. **hypo-** $(1)M_2S_2O_4$. (2) Thiosulfate. **sub-** Thiosulfate.
- s. ion.** The anion SO_3^{2-} . **s. cellulose liquor.** The waste liquor from the pulping of wood by the s. process: free SO_2 2-4, SO_3^{2-} 2-7, SO_4^{2-} 2-5, solids 80-90, ash 12-16%, with calcium compounds, carbohydrates, and lignin sulfonate. Used as a tanning, binding, or mordanting agent, or for fermentation. *s.c.l. lactone.* Tsugaresinol. **s. pulp.** Paper pulp obtained by the digestion of wood (generally coniferous) with calcium bisulfite containing free sulfur dioxide.
- sulfiviny.** The radical $=SO$. A term preferred to thionyl, except for the thionyl halides (U.K. usage).
- sulfo-, sulpho-** Prefix indicating the presence of (1) divalent sulfur, or (2) the sulfo group, $-SO_3H$. Cf. *thio-*. **s. acid.** Sulfonic acid. **s. amino.** Preferred term for *sulfamino*. **s. group.** The $-SO_3H$ group. See *sulfonic acids*. **s. salt.** (1) A salt of an acid containing sulfur. (2) An ester of sulfonic acid.
- sulfoacetic acid.** $HSO_3-CH_2-COOH \cdot H_2O = 158.10$. Colorless tablets, m.86 (sublimes), soluble in water.
- sulfoarsenide.** Sulfarsenide.
- sulfobenzide.** $(C_6H_5)_2SO_2 = 218.2$. Diphenylsulfone. Colorless scales, m.123, insoluble in water.
- sulfobenzoic acid.** $HSO_3-C_6H_4-COOH = 202.12$. **ortho-** Colorless, triclinic crystals, m.134 (forming an anhydride above 125), soluble in water. **meta-** Colorless crystals, m.141, soluble in water. **para-** Colorless needles, m.260, soluble in water. Cf. *halazone*.
- sulfobromophthalein sodium.** $C_{20}H_8O_{10}Br_4Na_2S_2 = 833.06$. White, hygroscopic bitter crystals; a diagnostic aid for liver ailments (U.S.P., B.P.).
- sulfocarbamide.** Thiourea.
- sulfocarbamilide.** $Ph \cdot NH \cdot CS \cdot NH \cdot Ph = 228.24$. Thio-carbamilide, diphenylsulfourea. Colorless leaflets, m.153, insoluble in water.
- sulfocarbazine.** A compound containing the radical $-NH \cdot NH \cdot CS \cdot NH \cdot NH-$.
- sulfocarbimide.** Isothiocyanic acid.
- sulfocarbodiazone.** A compound containing the radical $-N : N \cdot CS : N : N-$.
- sulfocarbonate.** A salt of phenolsulfonic acid.
- sulfocarbonic acid.** Phenolsulfonic acid.
- sulfocarbonate.** M_2CS_3 . A salt derived from sulfocarbonic acid.
- sulfocarbonic acid.** $H_2CS_3 = 110.2$. Trithiocarbonic acid. Brown oil, insoluble in water, readily decomposed; a reagent for nickel (as the K salt).
- sulfochloride.** $R-SO_2-Cl$. The chloride of sulfonic acids.
- sulfocyan.** Thiocyanate.
- sulfocyanate.** Thiocyanate.
- sulfocyanic acid.** Thiocyanic acid.
- sulfocyanide.** Thiocyanate.
- sulfoform.** $(C_6H_5)_3SbS = 384.8$. Triphenylstibine sulfide. Colorless needles, m.120; used to treat skin diseases. **crude-** Yellow liquid; a powerful reducing agent.
- sulfo group.** The $-SO_3H$ group. See *sulfonic acid*.
- sulfoguaiacin.** Quinine methyl sulfoguaiacate.
- sulfohydrate.** Hydrosulfide.
- sulfoichthyolic acid.** A compound obtained from bituminous shales; contains sulfur as sulfonate, sulfone, or sulfide.
- sulfoid.** Colloidal sulfur.
- sulfolane.** $C_4H_8O_2S = 120.26$. Tetramethylsulfone, tetrahydrothiophen-1,1-dioxide. Colorless solid, m.27, miscible with water; a selective solvent for hydrocarbons.
- sulfoleate.** A salt of sulfoleic acid.
- sulfoleic acid.** Obtained by mixing sulfuric acid with oils containing oleic acid.
- sulfolipins.** Fatty substances containing sulfur.
- sulfonal.** $Me_2C(SO_2Et)_2 = 228.3$. Sulphonal, sulfonmethane, diethylsulfonodimethylmethane. Colorless prisms, m.126, slightly soluble in water; a hypnotic. **methyl-** Trional.
- sulfonamic.** Sulfamic.
- sulfonamide.** Sulfamine.
- sulfonamido.** The radical $-NH-SO_2-$. Cf. *sulf-amino, sulfamyl*.
- sulfonaphthalein.** A compound similar to the phthaleins, but with a sulfone, instead of a carbonyl, group. Made by condensation of *o*-sulfobenzoic acid anhydride with phenols. Many are important indicators, q.v., and dyes; as, *o*-cresol s., cresol red.
- sulfonaphthol.** $HSO_3-C_{10}H_6-OH = 224.13$. Colorless powder; an antiseptic.
- sulfonate.** (1) To treat an aromatic hydrocarbon with fuming sulfuric acid. (2) A sulfuric acid derivative. (3) A sulfonic acid ester.
- sulfonation.** Substitution of H atom(s) by $-SO_3H$ group(s). **direct-** Treatment of an organic compound with fuming sulfuric acid. **indirect-** Treatment with acid sulfites.
- sulfonator.** A double-walled, cast-iron vessel with power-driven stirrer, for large-scale sulfonation.
- sulfone.** R_2SO_2 or $RSOOR$, obtained by oxidation of sulfides. In derivatives its prefix is sulfonyl*, Et_2SO_3 . **s. methanes.** Compounds used to induce sleep; as, trional, q.v. **s. phthalein.** See *sul-fonphthalein*.
- sulfonethylmethane.** Trional.
- sulfonic acid*.** An organic compound containing the radical $-SO_3OH$, derived from sulfuric acid by replacement of an $-OH$ group. Sulfonic acids are soluble in water and yield phenols when heated with potassium hydroxide. Used in the manufacture of dyes and synthetic drugs. **amino-** Sulfamic acid. **diamino-** Sulfamide. **nitro-** Nitrosyl sulfate.
- sulfonium.** Sulfine. The radical R_3S- . **s. compound.** R_3SX . Sulfine. R is an organic radical, and X an electronegative element (Cl) or radical

Hawley's
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Dictionary

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m-sulfobenzoic acid. $\text{HO}_3\text{SC}_6\text{H}_4\text{COOH}\cdot 2\text{H}_2\text{O}$.

Properties: Grayish-white solid. Stable but hygroscopic in air. Mp 98C, anhydrous form melts at 141C. Soluble in water, alcohol; insoluble in benzene.

Derivation: Direct sulfonation of benzoic acid with sulfur trioxide.

Use: Derivative for surface-active agents.

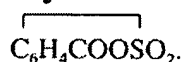
o-sulfobenzoic acid. $\text{HO}_3\text{SC}_6\text{H}_4\text{COOH}$.

Properties: White needles. Mp 68–69C (with $3\text{H}_2\text{O}$ of crystallization), mp 134C (dry). Soluble in water and alcohol; insoluble in ether.

Derivation: (1) From saccharin and concentrated hydrochloric acid, (2) by the oxidation of thiosalicylic acid with potassium permanganate in alkaline solution.

Use: Manufacture of sulfonaphthalein indicators, dyes.

o-sulfobenzoic anhydride.



Properties: Solid. Mp 129.5C, bp 184–186C (18 mm Hg). Soluble in hot water, ether, and benzene.

Use: Polymerization inhibitor.

sulfobetaines. See "Crosultaines" [Croda].

sulfocarbanilide. See thiocarbanilide.

sulfocarbolic acid. See phenolsulfonic acid.

sulfoderm. Silicic acid with 1% colloidal sulfur.

1-(4-sulfo-2,3-dichlorophenyl)-3-methylpyrazolone.



Properties: White or yellowish powder or crystals. Very soluble in water; soluble in alkalis.

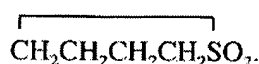
Derivation: By condensation of dichlorophenylhydrazine sulfonic acid with ethylacetoacetate.

Use: Intermediate for dyes.

"Sulfogene" [Du Pont]. TM for a series of sulfur colors.

Use: Extensively on cotton work clothing and similar fabrics and to a limited extent on rayon and other materials.

sulfolane. (1,1-dioxide tetrahydrothiofuran; tetramethylene sulfone).
CAS: 126-33-0.



Properties: Liquid. D 1.2606 at 30/4C, mp 27.4–27.8C, bp 285C, flash p 330F. Miscible with water, acetone, toluene (at 30C); slightly miscible with octanes, olefins, naphthenes.

Grade: Technical.

Hazard: Combustible. Toxic by ingestion.

Use: Curing agent for epoxy resins, medicine (anti-bacterial).

sulfonation. The introduction of the sulfo group into an organic compound whereby it connects to a carbon or nitrogen atom.

sulfonic acid. An organic compound containing in its molecule one or more sulfo radicals, $-\text{SO}_2\text{OH}$. The sulfur atom is directly united to a carbon atom.

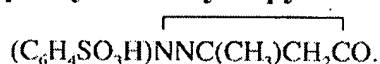
4,4'-sulfonyldiphenol. (SDP). $(\text{C}_6\text{H}_5\text{OH})_2\text{SO}_2$.

Properties: White, crystalline powder. Mp above 247C; 99.5% pure.

Derivation: Oxidation of 4,4'-thiodiphenol.

Use: Intermediate, product modification.

p-1-sulfohenyl-3-methyl-5-pyrazolone.



Properties: White or yellowish powder. Very slightly soluble in water; soluble in alkalis.

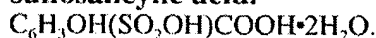
Derivation: By condensation of phenylhydrazine sulfonic acid with ethylacetoacetate.

4-sulfophthalic anhydride. $\text{HO}_3\text{SC}_6\text{H}_3(\text{CO})_2\text{O}$.

Properties: Reddish-brown syrup. Crystallizes partially on long standing; hygroscopic; fluorescent in solution under UV radiation. D 1.62 (25C). Very soluble in water, alcohol; insoluble in ether, benzene. Combustible.

Use: Esters of 4-sulfophthalic acid used in wetting, cleansing, emulsifying, softening, and equalizing agents with textiles. Derivatives have application as surface-active agents.

5-sulfosalicylic acid.



Properties: Colorless crystals, colored pink by traces of iron. Mp 120C (anhydrous), decomposes at higher temperatures. Very soluble in water.

Derivation: Action of sulfuric acid and salicylic acid.

Use: Reagent for albumin, colorimetric reagent for ferric ion, intermediate for dyes, surfactants, catalysts, grease additives.

sulfotepp. (ethyl thiopyrophosphate; tetraethyl dithiopyrophosphate).



Properties: Yellow liquid. Bp 136–139C (2 mm Hg). Slightly soluble in water; soluble in most organic solvents.

Hazard: Toxic by ingestion, inhalation, and skin absorption; cholinesterase inhibitor; use may be restricted. TLV: 0.2 mg/m³ of air.

Use: Insecticide.